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Review of Polymer Gels for Conformance Control in Oil Reservoirs

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By

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Review of Polymer Gels for Conformance Control in Oil Reservoirs

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ABSTRACT

Excess water production due to conformance problems is a serious issue in oil extraction with severe environmental and economic implications. This is mainly due to the heterogeneity of the reservoir and existence of thief zones which uptake the injected water. Polymer gels have been successfully used to improve the sweep efficiency and to mitigate excess water production. However, due to the complexity of the reservoir, reservoir temperature, salinity, pH, lithology, and permeability, selection of proper gel system is still challenging. This paper aims to provide a literature review on six widely applied polymer gel systems used for conformance control applications. For this purpose, various databases, such as Google Scholar, One-petro and Scopus were extensively searched. Results of this study reveal that polymer gel systems can mainly be classified into two categories: conventional in situ-bulk gels and novel microgels. The first type is mainly for water shut off near the wellbore, where a polymer in-situ cross-linked with a metallic or organic agents. The second type of gels include preformed gel particles with various sizes and properties which provide permeability reduction deep in the reservoir. This study summarized the characteristics, developments and field application results of six widely applied systems. Comparison of these technologies based on their properties and performance under different reservoir conditions is also provided. Directions for further research and development of these gel systems especially for improving their application in higher temperature reservoirs, extreme fractures and deep permeability reduction are given.

DEDICATION

This work is dedicated to

My beloved and Proud parents, ***Mrs. Shahla Gorbani*** *and* ***Mr. Torab Kazemi***

My beloved Sister, ***Sanaz***

who had always been the source of my inspiration.

&

My Proud Uncle ***Mr. Farhad Kazemi***

for being a continuous source of support, assistance and encouragement throughout this journey.

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I would also like to extend my appreciations to Dr. Xiaohong Xu for her guidance in the research and her generosity of granting me the Research Assistantship through her Natural Sciences and Engineering Research Council of Canada (NSERC) grant to assist me through my graduate study.

I would like to take this opportunity to thanks my committee member, Dr. Rajesh Seth for taking time to review my work and provide valuable advice and comments.

Finally, I would like to extend my appreciation to my family for their love, patience, understanding, and encouragement towards my achievements.

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LIST OF ABBREVIATIONS/SYMBOLS

Abbreviations

AM	Acryl-amide monomer
AMPS	Sulfonated acryl-amide monomer
BOPD	Barrels of Oil per Day
BP	British Petroleum (Company)
BWPD	Barrels of Water per Day
CDG	Colloidal Dispersion Gel
CIT	Conformance Improvement Technology
DPR	Disproportionate Permeability Reduction
EOR	Enhanced Oil Recovery
Gtoe	Giga tons of oil equivalent
HMTA	Hexamethylenetetramine
HPAM	Partially Hydrolyzed Polyacrylamide
HQ	Hydroquinone
IOR	Improved Oil Recovery
IFD	In-Depth Flow Diversion
OOIP	Original Oil in Place
OWC	Oil Water Contact
PAtBA	Polyacrylamide/tert-butyl acrylate
PAM	Polyacrylamide
PEI	Polyethylenimine
PPG	Preformed Particle Gel
PV	Pore Volume
RPM	Relative Permeability Modification
RPPG	Re-assembly Preformed Particle Gel
RSM	Rigid Settling Material
SMG	Smart Microgels
TAP	Temperature Activated Polymer
TDS	Total Dissolved Solid
WOR	Water to Oil Ratio
WSO	Water Shut-Off

Symbols

A	Swelling ratio
E_A	aerial sweep efficiency
E_D	microscopic displacement efficiency
E_I	macroscopic (volumetric) sweep efficiency
E_V	vertical sweep efficiency
e	adsorbed layer thickness
F_r	resistance factor
F_{rr}	residual resistance factor
K	permeability
K_v	vertical permeability
K_h	horizontal permeability
K_H	Huggins constant
k_{ro}	relative permeability to oil
k_{rw}	relative permeability to water
M	Mobility ratio
mD	milli-Darcy
R_F	Recovery factor
S_{or}	residual oil saturation
S_{wi}	residual water saturation
Q_o	oil flow rate
Q_w	water flow rate
λ_o	oil mobility
λ_w	water mobility
μ_w	water viscosity
μ_o	oil viscosity

CHAPTER 1

Introduction

The total primary energy supply in the world is projected to increase by 34.8% from 13.3 Gtoe in 2016 to 17.9 Gtoe in 2040 (BP energy outlook, 2018). The speed of energy transition from conventional to renewable sources of energy is uncertain. The evolving transition scenario which assumes that social preferences, technologies, and policies change with a pace similar to most recent years, predicts that the oil and gas sector contribution for energy supply will remain significant with the value of more than 50% of the total energy (BP energy outlook, 2018).With the global oil recovery factor of less than 34% and the difficulty in discovering of new oil fields, revitalizing and extending the life span of mature reservoirs become an important goals of the energy sector today (Abdulbaki et al., 2014; Ali, 2012).

The most widely used method to increase oil production is water-flooding (Mustoni et al., 2010; Alhuraishawy et al., 2017; Zaitoun et al., 2017). Water-flooding is the injection of water into the reservoir to displace the oil (Seright et al., 2006). Heterogeneity of the reservoir and existence of layers with high permeability (thief zones) restrict the effectiveness of water-flooding because water preferably passes through the layers with less resistance to flow. Therefore, existence of thief zones in the reservoir leads to impotent recirculation of water in the reservoir which consequently results in low oil recovery and excess water production (Imqam et al., 2018).

1.1 Problem Statement

Excessive water production is a significant challenge in the oil industry because it leads to unrecoverable oil in mature oil fields and has severe environmental and economic impacts (Mustoni et al., 2010; Alhuraishawy et al., 2017; Zaitoun et al., 2017). Controlling water flow in the reservoir during oil production has been the goal of the upstream oil industry (Bailey et al., 2000; Manrique et al., 2012). It is considered that the majority of the unwanted water production results from conformance problems that existed because of the heterogeneity of the oil reservoir (Thrasher et al., 2016; Bai et al., 2013). Polymer gels have been effectively used to address this problem. They are globally applied to improve the efficiency of water-flooding and other improved oil recovery (IOR) methods (Sydansk,

1990; Seright et al., 2006; Bai et al., 2007; Zaitoun et al., 2007; Al-Muntasheri et al., 2007). Polymer gels effectively block the high permeability thief zones and provide diversion of injected water toward low permeability un-swept zones. Such treatment of the conformance problems would cost-effectively extend the productive life of the reservoir by both mitigating the water production and recovering of bypassed oil reserves (Coste et al., 2000; Bai et al., 2013; Seright 2006a).

The selection of appropriate polymer gels for a specific reservoir is a difficult task for oil field operators. This is due to the complexity of conformance related problems that may encounter either near the wellbore or deeply in the formation. These systems were also prepared with various chemical properties, and forms. Furthermore, reservoir conditions, such as temperature, pH, salinity, degree of heterogeneity, and type of rocks are also complicating the application of these technologies. Therefore, the success and effectiveness of treatment highly depend on the proper selection of the system. Various polymer gel systems have been introduced in both oil fields and laboratory experiments over the past five decades to address various conditions encounter during the treatment process.

Several authors reviewed various polymer gel systems used for the conformance control application over the past two decades (Moradi, 2000; Vossoughi, 2000; Vasgas-Vargas-Vasquez and Romero-Zeron, 2008; Chung et al., 2011; Han et al., 2014; Abdulbaki et al., 2014; El-Kasrani et al., 2014b; Bai et al., 2015; Zhu et al., 2017; Amir et al., 2019; Ghriga et al., 2019). The most recent reviews were focused on the review of development of polymer gel systems for in-depth flow diversion application (Chung et al., 2011; Abdulbaki et al., 2014; Bai et al., 2015) and polymer gel systems for high temperature and high salinity reservoirs (Zhu et al., 2017; Amir et al., 2019; Ghriga et al., 2019).

Vargas-Vasquez and Romero-Zeron (2008) provides a review on the factors affecting HPAM/Cr (III) acetate gelation kinetic, gelation time, gel strength, gel stability, syneresis and rheology. Abdulbaki et al. (2014) gave a review of four different polymer microgels for in-depth flow diversion applications. Four different types of microgels including colloidal dispersion gels, preformed particle gels, temperature activated microgels and pH-sensitive polymers were reviewed in their paper. Their review covers

the characteristics of four types of microgels with the focus on both lab and field studies. El-Kasrani et al. (2014b) provided a review of the polymer gel systems introduced between the years 2001 to 2011, regardless of being implemented in the oil field or introduced in the lab experiments. Bai et al. (2015) presented a thorough review of polyacrylamide based gel systems and based on their form classified them into three categories: in-situ monomer based, in-situ polymer based and preformed gels. They also compared these three categories based on their ability to provide deep flow diversion. Zhu et al. (2017) reviewed polymer gel systems technologies and categorized them into three groups: in-situ cross-linked, foamed gels and preformed gels. This study covers a large number of polymer gel systems with various chemistries that have been introduced in the lab and field for high reservoir temperature water management applications. Most recently, Amir et al. (2019) with the same purpose provide a literature review on the organically cross-linked in-situ polymer gels for high salinity and high temperature reservoirs. Different organically cross-linked in-situ gel systems are discussed in terms of chemistry and gelation kinetics. Factors affecting the gelation time of the gel systems are also extensively reviewed. In their review, they covered phenol based, formaldehyde based and polyethylenimine cross-linked gel systems. Ghriga et al. (2019) specifically focused on the review of polyethylenimine based organically cross-linked gel systems for high temperature reservoirs. In their review, they studied various polymers/PEI gel systems, the lab and field observations regarding their gel strength and gelation time of these systems are reviewed.

Among all polymer gel systems that are studied for the past two decades, six polymer gel systems including HPAM/Cr (III) acetate, PAtBA/PEI, CDGS, PPGs, TAPs and Microgels are commercialized and widely applied around the globe. Table 1-1 shows the summary of previous reviews that covered one or some of these widely applied polymer gel systems. Although, most of these reviews studied some of these polymer gel systems but based on my best knowledge there is no document that fully covered all these six widely applied technologies. The previous reviews that covered most of these systems were focused mainly on chemistry, kinetic and gelation time and gel strength through lab and field observations of these systems. However, other aspects of polymer gel systems such as relative permeability modification, selectivity of penetration, in-depth permeability reduction and methods used in field or lab to improve the performance are not fully

covered. Furthermore, for the past five years (2015-June 2019), there are substantial number of papers published on the development and characteristics of these six technologies that are not addressed in previous review studies.

Table 1-1: Overview of previous review papers on polymer gel systems.

Author and Year	Systems covered	Focus of the study	Factors Considered
Vargas-Vasquez and Romero-Zeron, 2008	HPAM/Cr(III) acetate	Factors affecting cross-linking reaction kinetic, rheology, gelation time, gel strength, syneresis, gel stability	Temperature, Solvent salinity, Cross linker concentration, reservoir minerals, polymer hydrolysis, polymer molecular weight, shear environment, polymer concentration
Abdulbaki et al., 2014	CDGs, PPGs, TAPs, pH-sensitive polymers	Review of polymer microgels for conformance control	Microgels characteristics, laboratory observations, field applications, rheology and plugging mechanism.
El-kasrani et al., 2014	PAM/PEI, AMPS/PEI, PHPA/Chitosan, Polyurethane Resin, PAtBA/PEI, PDVSA Gel, AMPS/N,N'-DMA/PEI PAtBA/Chitosan, PHPA/Cr^{+3} Foam, PHPA/ Cr^{+3} Nano-particles, PHPA/terpolymer Cr^{+3}, CDGs, PPGs, TAPs	Review of development of polymer gel systems for deep modification of water injection profile and near wellbore water shutoff between years 2001-2011	Highlight advances , developments advantages, shortcomings and summarized the field applications
Bai et al., 2015	In-situ monomer gels, in-situ polymer gels including HPAM/Cr (III) acetate & PAtBA/PEI, preformed gels including CDGs,PPGs,TAPs	Review the development of Polyacrylamide based polymer gel systems based on their composition, form and application condition	Chemistry, characteristics, advantages, disadvantages, field applications
Zhu et al., 2017	In-situ gel systems including PAtBA/PEI Preformed gels including TAPs and Microgels and Foam gels	Chemically review the polymer gel systems for high temperature and high salinity reservoirs	Gelation formulation, gelation time, gel strength

Amir et al., 2019	Phenol-formaldehyde cross-linker Hexamethylenetetramine cross-linker Polyethylenimine cross-linker including PAtBA/PEI	Study of organically cross-linked systems for high temperature reservoirs in terms of chemistry, gelation mechanism , factors affecting gelation kinetics and field application	Temperature, initial pH, Salinity, Polymer concentration, Cross-linker concentration, additives
Ghriga et al., 2019	PAtBA/PEI PAM/PEI PHPA/PEI HAP/PEI Other polymers/PEI	Highlight recent improvement of gelation time and gel strength of polymer/PEI systems	Chemistry, gelant composition, gelation kinetics, Effect of additives

* Explanation of bold terms used in second column of the Table 1-1 can be found in List of ABBREVIATONS (page xii).

1.2 Objectives of the Research

This study aims to provide a review of the polymer gel technologies that are commercialized and widely applied in the oil fields. The overall objective of this research is to provide an updated review on the six widely and globally applied polymer gel technologies in the area of conformance control. The specific objectives are to:

1) review the characteristics, development and application of most widely applied polymer gel systems.
2) compare the selected technologies based on their properties and their performance at reservoir conditions.

This study provides an updated review that summarizes results of the previous field treatments and lab observations, which is helpful to the reservoir engineers and oil field operators. It will also provide them with the methods that have been used to further improve the effectiveness of these technologies. Finally, the review provides new insights about these polymer gel technologies, identifies the gaps in the literature, and provides directions for future research of polymer gel systems improvement for various conformance and reservoir conditions.

1.3 Methodology

Similar to most literature review studies, the internet and in particular Google Scholar search tool and University of Windsor collections were used to conduct the research. The first step was to search for "Conformance Control" and "Polymer Gel" phrases targeting scientific journals, theses, and dissertations. With identifying the scope of the research, six polymer gel systems include HPAM/Cr (III) acetate, PABA/PEI, CDGs, PPGs, SMG Microgels, and TAPs were selected for further research. For this purpose, major keywords such as "HPAM", "Chromium", "PAtBA", "PEI", "CDGs", "PPGs", "TAPs", "Bright Water", and "Microgels" combined with phrases such as, "Conformance Control", "Water shutoff", "Profile Modification" and "In-Depth Flow Diversion" were used for further searches. A variety of databases were searched, including Google Scholar, One Petro, Scopus, Science Direct, Wiley Online Library, and ProQuest. A quick review of the research results indicated that the most valuable sources are coming from the Society of Petroleum Engineering (SPE) peer-reviewed journals and conference publications. Polymer gels have a longtime application in conformance control; thus, the initial search has returned a substantial number of results. The search revealed over 700 papers that were related to the topics. As a result, a need for filtering procedure to keep the most valuable resources became more apparent. The main inclusion/exclusion criteria were specialization, originality, and research date as detailed below:

(a) Specialization: include the publications which were specific to the application of polymer gels for conformance control that are commercialized and widely applied in the oil fields. Exclude the publications which were related to application of polymer gels for other purposes such as well abandonments and polymer gels systems that have not been implemented in field applications.

(b) Originality: selected innovative, new and unique studies from peer-reviewed and conference publications which were resulted from field observation and lab experiments. Also, using different databases, such as Google Scholar, an effort has been made to find the most cited publications in the previous literature review documents.

(c) Research duration and language: the publications were limited to English, and the publications from the time period of 2015-June 2019 were prioritized. The outdated research papers which were about obsolete technologies were excluded.

The identified literature was reviewed, and papers were chronologically and thematically categorized. The combination of data analysis including inclusion/exclusion criteria and sorting the relevant information in different categories, has led to accomplish a framework for obtaining the valuable information and knowledge about the research topic and perform the required analysis.

1.4 Brief Description of Chapters

Chapter 2 focuses on the reviews of the concepts of enhanced oil recovery, conformance control, and the relation between reservoir conformance and excess water production. Conformance issues causing poor recovery and excess water production are discussed in some details. Finally, conformance improvement technologies, including mechanical, completion, and chemical methods, are explained briefly.

Chapter 3 discusses the application of polymer flooding and polymer gel treatments. The commonalities and differences between these two technologies are explained. The standard terms used to measure the performance of these technologies quantitatively are described. Because the main focus of this research is on polymer gel treatments, different types of gel treatments terms and operations are explained.

Chapter 4, as the main body of this study, reviews the literature on the six commercially available polymer gel technologies in conformance control applications. Polymer gel systems were categorized into two main groups i.e., conventional in-situ bulk gels and novel microgels. For each of the selected technologies, both field application results and relevant laboratory experiments are reviewed. The introduction to the technologies, their development, the effect of reservoir condition (temperature, salinity, pH, etc.) and other important information related to the performance of these technologies are explained and summarized.

Chapter 5 presents the conclusions. Some recommendations for further research and improvements are provided as well.

CHAPTER 2

Enhanced Oil Recovery and Conformance Issues

2.1 Enhanced Oil Recovery and Conformance

Primary, secondary and tertiary recoveries are the three main stages of oil production (Sheng, 2011). In the primary recovery, the flow of the hydrocarbon into the wellbore is induced by using pumps (artificial lift) and by mechanisms that are naturally occurring in the reservoir (Sheng, 2011). Such mechanisms include water drive, gravity drainage, gas cap drive, solution gas drive, and fluid/rock expansion (Sheng, 2011). The primary recovery stage is not economically viable in the long term, because it is able to recover only up to 15% of the original oil in place (OOIP) (Green and Willhite, 1998).

Secondary recoveries involve the injection of immiscible fluid, gas or water (water flooding) in order to maintain the reservoir pressure and to displace the remaining oil in the reservoir, and they can produce an extra 10 to 15% of original oil in place (OOIP) (Green and Willhite, 1998; Sheng, 2011). Generally, primary and secondary recovery can account for the extraction of approximately, 35% of the total oil in the reservoir (Green and Willhite, 1998).

Tertiary oil recovery or the so-called enhanced oil recovery (EOR) technique is applying energy or chemicals that do not naturally exist in the reservoir to extract the remaining oil in the reservoir after primary and secondary recovery (Green and Willhite, 1998). Thermal recovery, chemical injection and miscible (gas) injection are the three main categories of EOR methods (Green and Willhite, 1998). Thermal recovery furthermore divided into in-situ combustions, steam flooding, cyclic steam injection, and steam aided gravity drainage (SAGD) (Sheng, 2011). Chemical injection (flooding) methods include surfactant flooding, alkaline flooding, polymer flooding, and microbial methods (Sheng, 2011). The miscible injection methods include nitrogen flooding, carbon dioxide flooding, cyclic carbon dioxide simulation and solvent flooding (Sheng, 2011; Sydansk & Romero, 2011). Figure 2-1 summarizes the recovery methods and also another IOR method, conformance control. Conformance controls are not oil recovery methods but they widely used to improve the performance of secondary and tertiary methods. The concept of

conformance control and its role on improving the oil recovery are explained in more details in the following sections of this chapter.

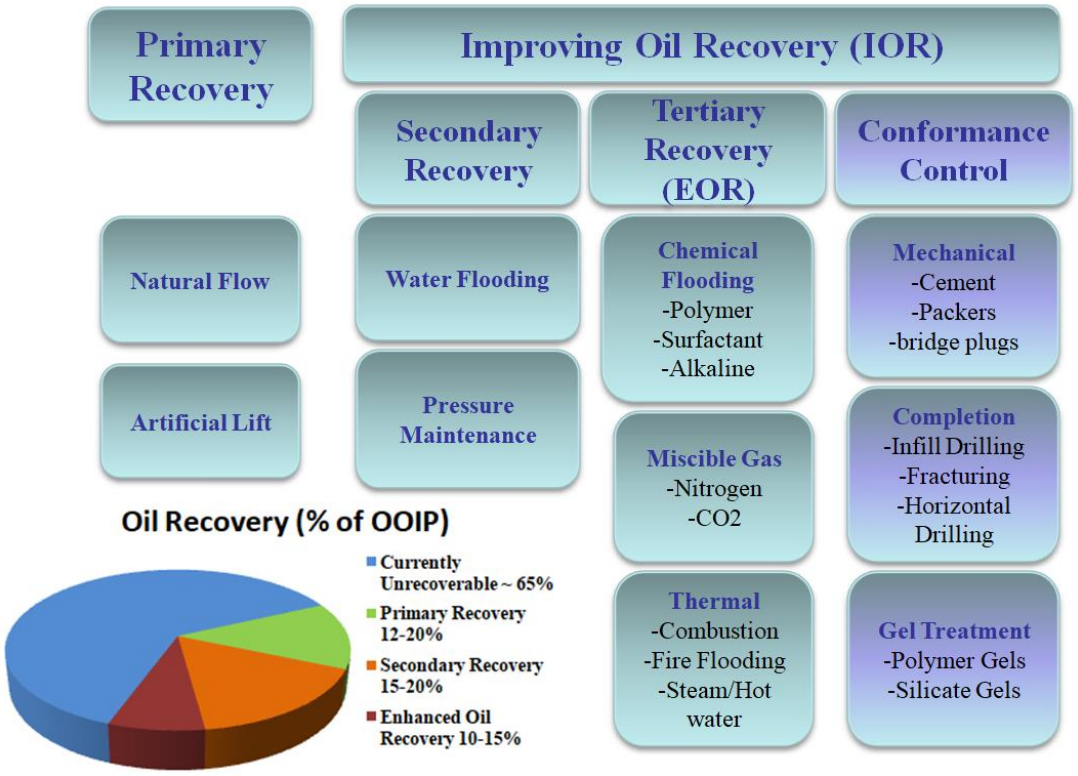


Figure 2-1: Enhanced oil recovery methods.

For any of the recovery methods mentioned in Figure 2-1, the total recovery factor (R_F) is defined as the product of macroscopic displacement or volumetric sweep efficiency (E_I) and the microscopic displacement efficiency (E_D) as follow:

$$R_F=E_D\times E_I \tag{2.1}$$

The volumetric sweep efficiency (E_I) is defined as the product of areal (E_A) and vertical sweep efficiency (E_V):

$$E_I=E_A\times E_V \tag{2.2}$$

Equations (2.1) and (2.2) indicate that the improvement of the oil recovery can be achieved by the improvement of both microscopic and macroscopic efficiencies (Green and Willhite, 1998; Sydansk & Romero, 2011).

The microscopic displacement efficiency (E_D) is defined as the volume of oil removed from the swept zones for any pore volume of the injected fluids (Sydansk & Romero, 2011). The microscopic displacement efficiency is related to the residual oil saturation (S_{or}), or oil remained in the area of the reservoir that is already swept (Sydansk & Romero, 2011). The presence of capillary force, viscous force, rock wettability, interfacial tension and surface tension between fluids and rocks in the reservoir are the factors that are controlling the residual oil saturation (Green and Willhite, 1998).

Generally, displacement efficiency, is improved when oil viscosity, capillary force, and interfacial tension decreased and the rock becomes water wet (Green and Willhite, 1998). Therefore, this efficiency can be improved with the injection of any material that can target the rocks and fluids interactions. For example, in the case of surfactant flooding, the mechanism of oil displacement is based on the reduction of interfacial tension, while polymer flooding increases the displacing fluid viscosity (Green and Willhite, 1998). In the case of steam injection, the heat applied to the oil reduces the viscosity of the oil and improves the displacement, and solvent injection helps the oil remained in the pores to move easier by reducing the capillary force. Alkaline flooding proved to be effective in enhanced oil recovery by reducing interfacial tension and wettability alteration (Sydansk & Romero, 2011).

Volumetric sweep efficiency (E_I) is defined as the percent of the pore volume that is swept by the injection fluid to the total volume containing oil (Sydansk & Romero, 2011). Figure 2-2 illustrates the most important reservoir poor recovery reasons. Poor volumetric sweep efficiency in an oil reservoir can be due to the following reasons:

- Heterogeneity of the reservoir causes the displacing fluid to flow through areas/zones of high permeability.
- Fractures in the reservoir.
- Viscosity of the displacing fluid is less than oil and can cause viscous fingering of the injected fluid.
- Oil wet rock

Therefore, the volumetric sweep efficiency (E_I) can be improved by modifying the permeability, wettability alteration, decreasing oil viscosity or increasing displacing fluid viscosity (Sydansk & Romero, 2011).

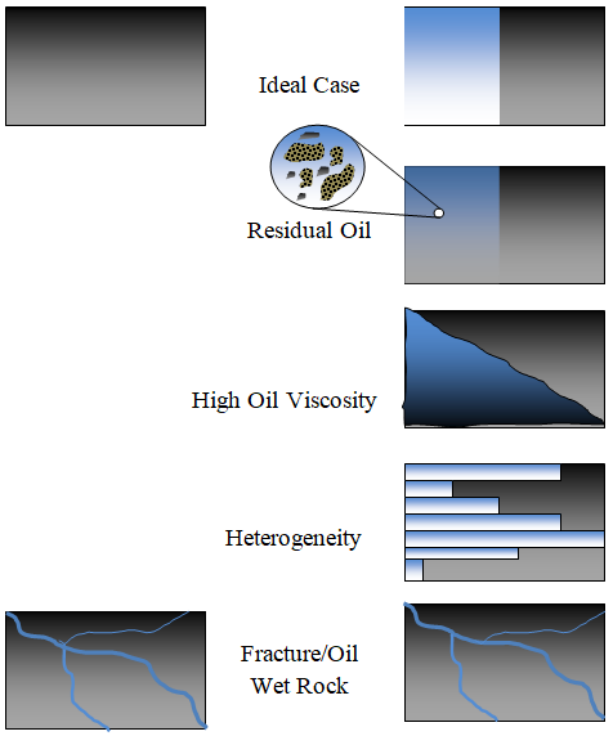


Figure 2-2: Five main reasons for reservoir poor recovery (adopted from Green and Willhite, 1998).

In general, the measure of volumetric sweep efficiency during any oil-recovery process that conducts flooding is described with the term conformance (Sydansk & Romero, 2011). The term conformance also used widely to address the excessive water production during oil recovery. It is clear that excess water production and early water breakthrough has a negative impact on overall volumetric sweep efficiency and oil production (Sydansk & Romero, 2011). To visualize favorable conformance two premises should be kept. First, the displacing fluid contacts the oil bank in every region in the reservoir and second, the oil recovery flood front moves easily and equally throughout the whole volume of the reservoir (Sydansk & Romero, 2011).

Figure 2-3 illustrates the ideal conformance and aerial and vertical conformance problems. As Figure 2-3 (a) shows, the non-uniform aerial and vertical flood front or so-

called conformance problems are caused by unfavorable mobility ratio and/or heterogeneity of the reservoir which referred to as conformance problem roots. In Figure 2-3 (a) on the vertical view, the layer three with the higher permeability than other layers (K3>K2>K1>K4) would uptake the water while the oil in the layer four with the lowest permeability remains un-swept. The mobility ratio (M) greater than unity means water has higher mobility than oil and as shown in aerial view of Figure 2-3 (a) the injected water finds its way to the produced with fingering and oil remains un-swept. Figure 2-3 (b) shows an ideal case where mobility ratio (M) is less than unity and all four layers having same permeability (K1=K2=K3=K4). In this case, all the volume of the reservoir would sweep by water and results in more oil recovery.

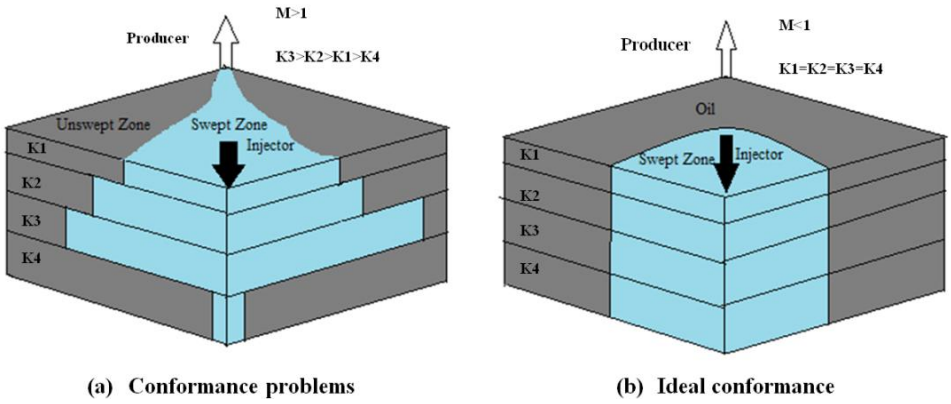


Figure 2-3: Reservoir conformance, (a) poor and (b) ideal.

2.2 Excessive Water Production

Oil production is usually accompanied by water production (Lantz and Muniz, 2014). Excessive water produced during oil and gas operation is an issue that is affecting all of the oil reservoirs worldwide (Bai et al., 2013). The produced water reduces the expected economic life of the reservoir and creates significant technical and environmental problems (Imqam et al., 2017). As reservoir undergoes water-flooding and becomes mature, the issue of water production increases (Bai et al., 2013). As reservoir matures and undergoes water-flooding, the water can be as much as 98% of the material extracted (Yusta-García et al., 2017).

The common term used by oil operators to address this problem is water to oil ratio (WOR). WOR defined by Equation 2.3 as follow:

$$\mathrm{WOR} = \frac{\mathrm{Qw}}{\mathrm{Qo}} \tag{2.3}$$

where Q_w and Q_o represent the flow rates of water and oil, respectively (Sydansk & Romero, 2011).

It is reported that on average three barrels of water are produced for one barrel of oil on the global scale (Bailey et al., 2000). However, in the United States, the average WOR is reported to be around eight (Al-Muntasheri, 2012). The oil and gas industry produced an average of 33.4 million m^3 of water each day in 2000, and this value increased to 39.64 million m^3 in 2005 (Al-Muntasheri, 2012). In the North Sea oil reservoirs, the problem was worse, where 222 million tons of water was produced each day for only 4 thousand tons of hydrocarbon (Al-Muntasheri, 2012). Van Eijden et al. (2004) reported that the water production in the Shell group has increased substantially from 2.2 million barrels/day to more than 6.3 million barrels/day in less than 15 years.

Cost of handling, lifting, de-oiling, pumping, separation and disposal of large amount of water; increased rate of corrosion, scaling and sand production; environmental concerns and liabilities; and, damage to formation by re-injection are among the main problems associated with early water breakthrough which often impose additional costs to the production and significantly impact the ultimate recovery (Seright et al., 2003).

Bailey et al. (2000) estimated that the average annual cost of disposal of produced water worldwide was $40 billion in 1990 and this amount was reported to be $42 billion in 2002 (Bøye et al., 2011). Hill et al. (2012) mentioned that the annual cost of separations, disposal, and treatment of produced water in the global scale was $ 50 billion. The most recent analysis on produced water treatment market (Grand View Research Group, 2016), shows that the strict environmental regulations progressively increased the treatment market size. The cost of excessive water treatment in 2015 was USD 5.81 billion and expected to reach USD 9.8 billion by 2024 (Grand View Research Group, 2016).

2.3 Sources of Water Production Problems

Water production problems can be categorized into two main groups based on their proximity to the wellbore: (1) Near-wellbore related problems, and (2) Reservoir-related problems (Bailey et al., 2000).

2.3.1 Near Wellbore Problems

Near wellbore problems usually take place during the early stages of oil production and are from either mechanical or completion roots (Bailey et al., 2000).

Mechanical problems

If any of the casing, tubing or packer has poor mechanical integrity, the leakage of water is likely to occur. The failure may be due to the corrosion of the casing or excessive pressure during operations. As Figure 2-4 shows, the leaks allow water to penetrate into the wellbore from water zones bellow perforation (Bailey et al., 2000).

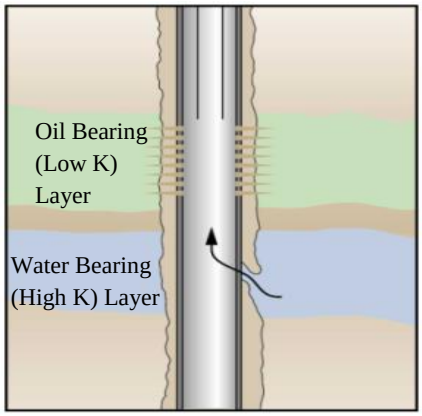


Figure 2-4: Casing, tubing or packers leaks (Bailey et al., 2000).

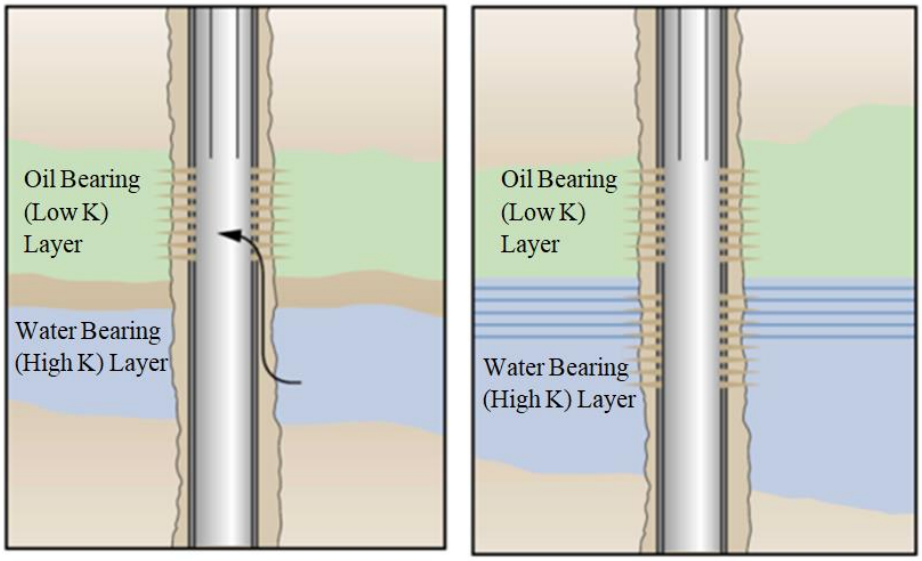
Completion problems

The two most common completion problems are (1) flow behind casing, and (2) moving oil-water contact (Bailey et al., 2000).

Flow behind casing Inadequate or failed primary cementing can connect water-bearing layers to the perforated zone. As Figure 2-5 (a) shows, these roots allow the water to flow into the annulus (Bailey et al., 2000).

Moving oil-water contact During normal water-driven production in a well a uniform oil-water contact might move up to the perforated zone and lead to unwanted water

production (Figure 2-5 (b)) This type of problem may occur when the oil water contact (OWC) and perforations are close to each other and there is a low vertical permeability in the formation (Bailey et al., 2000).



(a) Flow behind casing (b) Moving OWC

Figure 2-5: Completion problems, (a) flow behind casing and (b) moving oil water contact (Bailey et al., 2000).

2.3.2 Reservoir Related Problems

These types of problems are usually occurring when a reservoir matures or at least has gone through some production. The water production problems are mainly due to the permeability heterogeneity of the reservoir and/or viscosity contrast between water and hydrocarbons (Bailey et al., 2000).

High permeability layer without cross-flow Figure 2-6 shows a high permeability layer between two shale layers. Shale layers with having very low permeability are working as barriers. In this case, the water source may be from a water flood injection well or an aquifer. Because there is no pressure communication between layers, water preferably flows through the high permeability zone (Bailey et al., 2000).

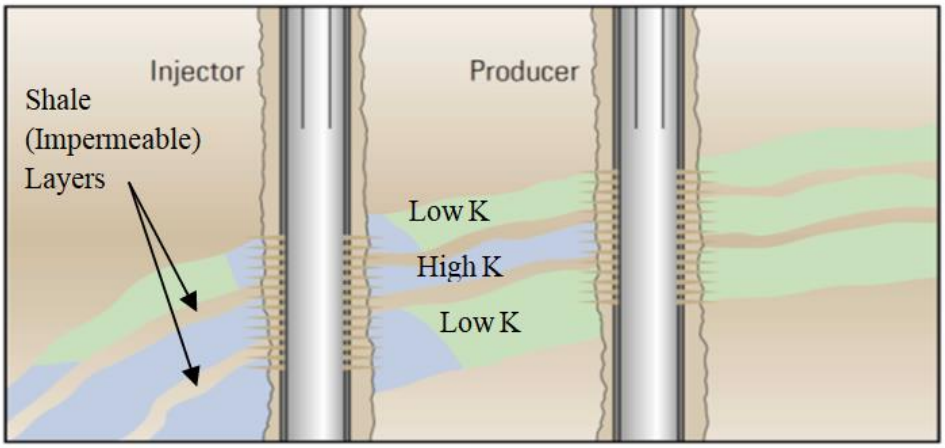


Figure 2-6: High Permeability layer without cross flow (Bailey et al., 2000).

Fractures between injector and the producers in naturally fractured formations such as carbonate reservoirs, as shown in Figure 2-7, the injected water can easily and rapidly breakthrough if there is a fracture that connects the two wells (Bailey et al., 2000).

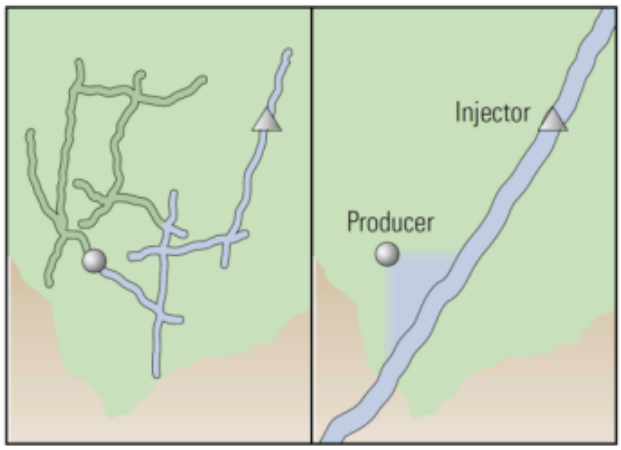


Figure 2-7: Fractures between injector and the producer (Bailey et al., 2000).

Fractures from a water layer Figure 2-8 shows how natural fractures in the water-bearing zones can contribute to the water production. This type of problem can also initiate after hydraulic fracturing if the fractures penetrate to the water-bearing zone on top or bottom of the oil-bearing zones (Bailey et al., 2000).

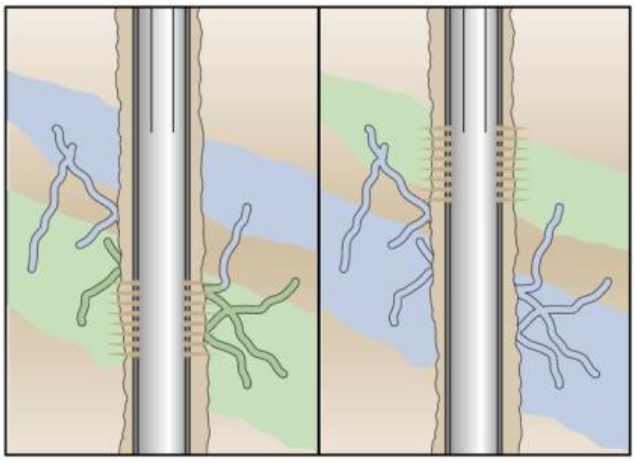


Figure 2-8: Fissure/Fractures from water zone (Bailey et al., 2000).

Conning and Cusping Figure 2-9 shows the conning problem in vertical well and similar problem in a horizontal well, the cusping. These types of problems occur when the OWC (oil-water contact) and perforations are close to each other, and there is a high vertical permeability in the formation. As the production rate increased the water below OWC move upward because of high vertical permeability, the minimum rate at which water starts to produce in this case is called critical conning rate.

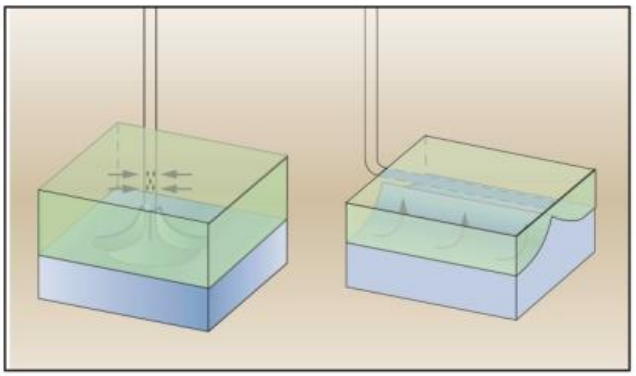


Figure 2-9: Conning and cusping (Bailey et al., 2000).

Poor areal sweep Figure 2-10 shows water flooding through a layer with poor areal sweep and edge water from an aquifer. These are usually due to adverse mobility ratio or areal permeability heterogeneity. Poor sweep efficiency is a more common problem in formations with sand channels (Bailey et al., 2000).

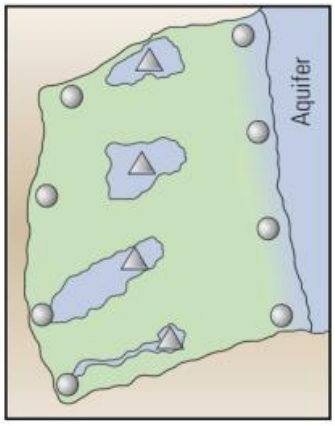


Figure 2-10: Poor aerial sweep (Bailey et al., 2000).

Gravity segregated layer This type of water problem is common for thick layer of reservoir with having vertical permeability. It is also referred to as water under run. As shown in Figure 2-11, the water from the water flood, sweep only the lower part of the formation and cause excessive water production at the producer. The main reason for this problem is due to the higher density of water compared to oil, and the problem even gets worse if the oil has relatively higher viscosity than water (Bailey et al., 2000).

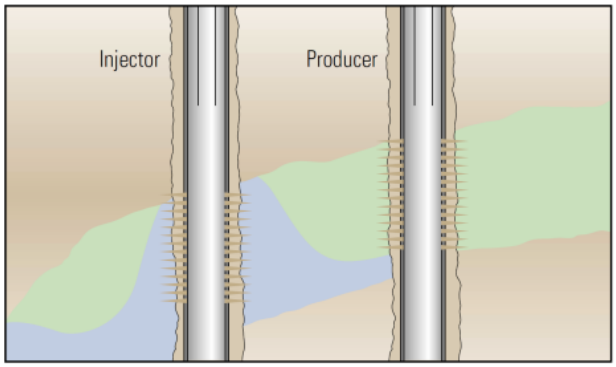


Figure 2-11: Water under run (Bailey et al., 2000).

High permeability layer with cross-flow Figure 2-12 shows a high permeability streaks similar to Figure 2-6 but there are no shale layers as barriers for cross flow of water between adjacent zones. Layers in the reservoir are in pressure communication. These types of problems are more difficult to treat because the treatment needs to be applied deep into the formation (Bailey et al., 2000).

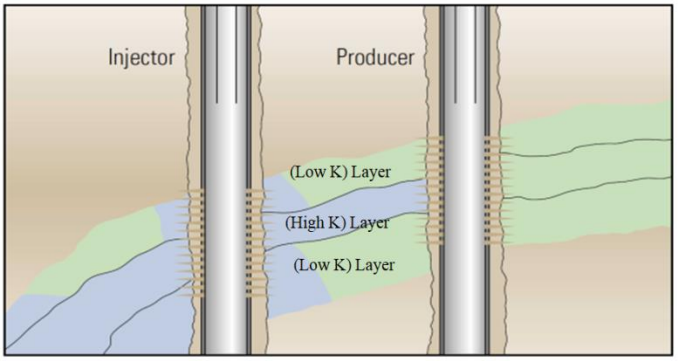


Figure 2-12: High permeability layer with cross-flow (Bailey et al., 2000).

2.4 Conformance Improvement Technologies (CITs)

Conformance improvement technologies (CITs) are available technologies to enhance the efficiency of IOR methods such as water-flooding and to tackle the excessive water production problem in oil reservoirs (Seright et al., 2003). Seright et al. (2003) categorized the conformance solutions into conformance agents and conformance practices/operations as shown in Table 2-1. The first category includes any chemical or physical materials that can be injected into the reservoir as a plugging agent (Seright et al., 2003). For instance, polymers, polymer gels, resins, and cement can be injected near the wellbore or far into the reservoir to block a layer or change the permeability disproportionally. The second category includes completion or mechanical techniques such as infill drilling, hydraulic fracturing, using packer and bridges (Seright et al., 2003).

Table 2-1: Conformance improvement materials and techniques (regenerated with permission from Seright et al., 2003).

Conformance Agents	Conformance Operations
• Foams, emulsions, particulates Precipitates, microorganisms • Cement, Sand, Calcium carbonate • Resins • Polymer/mobility-control floods • Polymer Gels	• Packers, bridge plugs, patches • Well abandonment • Infill drilling • Pattern flow control • Horizontal wells, advanced wellbore • Fracturing

Conformance solutions can also be categorized as mobility control and conformance control methods based on their objectives (Sydansk & Romero, 2011). If the technology tries to solve the problem related to the viscosity or density differences between the drive-fluid and oil, the method is hereafter referred to as mobility control (Sydansk & Romero, 2011). On the other hand, the technologies that are trying to improve the production and/or injection profile are referred to as conformance control methods (Sydansk & Romero, 2011). These types of conformance improvement technologies try to correct the reservoir permeability heterogeneity and consequently improve the production by enhancing the sweep efficiency of the flooding process (Bailey et al., 2000; Sydansk & Romero, 2011). In the following subsections the conformance control treatments are categorized as mechanical, completion and chemical methods and explained in more details.

2.4.1 Mechanical Methods

Mechanical methods usually refer to the use of hardware, such as bridge plugs, straddle packers, tubing patches, water separation tools or cement to shut off water flow (Bailey et al., 2000). These methods are often used to address near wellbore issues, such as flow behind pipes, casing leaks, rising bottom water and in some case for high permeability streaks if there is a no cross-flow between reservoir layers (Bailey et al., 2000). Figure 2-13 shows the application of mechanical plugging tools for water shut off near the wellbore. As shown in Figure 2-13, if there is a shale layer between oil zone and water zone, then setting a mechanical tool can be useful to plug the flow of water from the water-out zone into the wellbore.

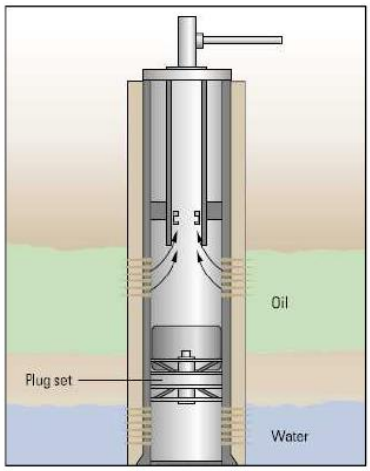


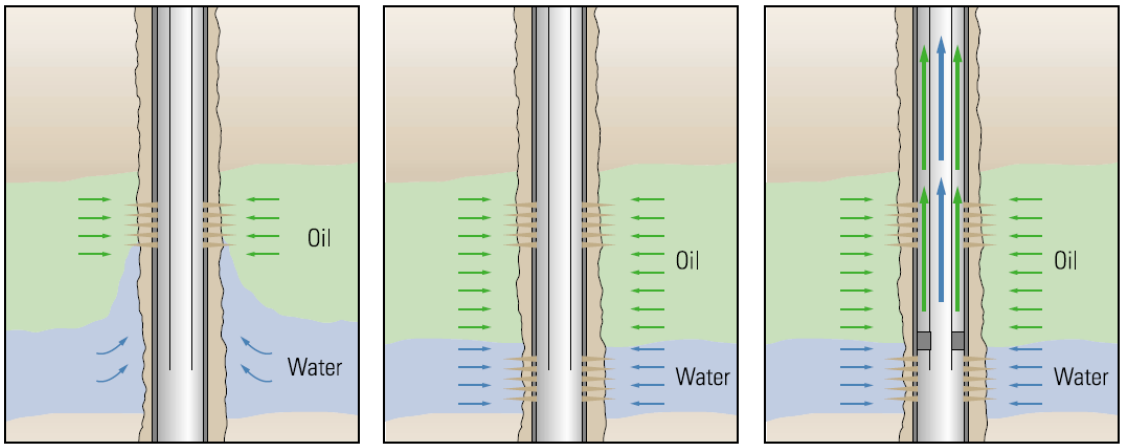
Figure 2-13: Mechanical plug tools (Bailey et al., 2000).

However, the mechanical methods are not enough to solve all the excessive water problems. There are some limitations for mechanical methods. First, in problems, such as flow behind pipes or casing leaks, the aperture sizes are usually smaller than the particle size of the sealing material which makes the penetration of the sealing material impossible and ineffective. Another problem is the damage of the mechanical methods to the formation especially damages to oil pay zones because of lack of control. Generally, there is very low control over these types of methods. Also, the mechanical methods usually require work-over rig and therefore are expensive. In cases where the problem is reservoir related rather than near the wellbore cement penetration deep into the formation also has some limitations because cement might not be placed in the targeted zone and consequently damage the hydrocarbon zones (Seright et al., 2003; Bailey et al., 2000).

2.4.2 Completion Methods

Completion methods, such as dual completion, sidetracks, coiled tubing isolation, and multilateral wells can be implemented to solve some of the problems related to water production such as incomplete areal sweep, gravity segregation, and 3D conning. Figure 2-14 illustrates an example of a water problem solved with the completion method. In this case, a water conning problem is solved with dual completion and re-perforation (Bailey et al., 2000). As shown in figure 2-14 water from the high permeability layer can move up to the perforation area if there is a high vertical permeability (K_v). Figure 2-14 (b) shows the re-perforation of the water bearing zone of the reservoir, which prohibited the water

from conning. Figure 2-14 (c) shows the application of packers and dual completion, which results in separate production of oil and water. This remediation not only treats the conning of water but also significantly reduce the cost of water treatment because there is no need for de-oiling and separation processes.



(a) water conning (b) re-perforation (c) packers and dual completion

Figure 2-14: Re-perforation and dual completion method for conning problem (Bailey et al., 2000).

2.4.3 Chemical Methods

Generally, mechanical and completion solutions to water production problems are referred to as conventional methods (Seright et al., 2003). Although mechanical and completion solutions can be used to solve some of the wellbore as well as near wellbore problems, some conformance problems need to be treated with the penetration of material deep into the reservoir or required small fissures penetration ability of the sealing materials (Seright et al., 2003). Because of the weak points mentioned above, there is a need for a material with a good level of penetration and sealing. These properties are available in some chemicals including resins, foams, emulsions, polymers, and gels (Liu et al., 2006). Polymers and polymer gels are the most widely used chemical materials in the area of conformance improvement technology (Bai et al., 2015). Because of the importance of polymer and gels in conformance improvement and to differentiate their applications from each other; the next chapter is specifically devoted to these two technologies.

CHAPTER 3

Gel Treatment

3.1 Polymer Flooding and Gel Treatment

Polymer is one of the widely used chemical materials in enhanced oil recovery and conformance improvement (Sang et al., 2014). Sorbie and Seright (1992) differentiated polymer gel treatment from conventional polymer flooding. As shown in Figure 3-1(b), in gel treatment, the goal is to minimize the penetration of the gel or gelant into the low permeability oil-rich zone and maximized the penetration in high permeability water out zone. While, as Figure 3-1(a) shows, in traditional polymer flooding, the polymer penetration in the low permeable oil-rich zone should be maximize and the injection in high permeable and already sweep water zone should be minimized (Sorbie and Seright, 1992).

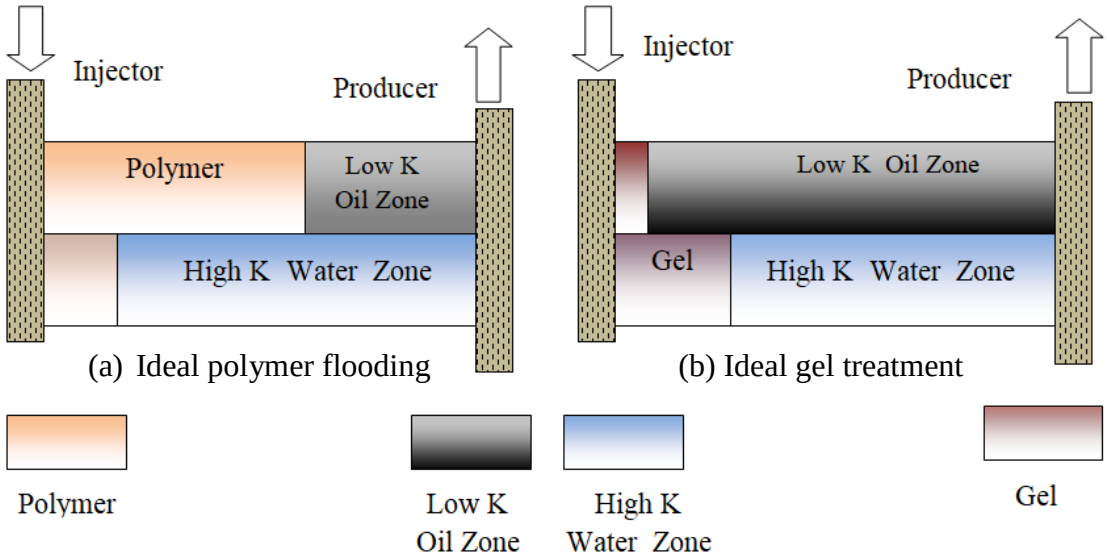


Figure 3-1: (a) Ideal polymer flooding and (b) ideal gel treatment.

In general, gel treatment has different applications than traditional polymer flooding (Sorbie and Seright, 1992). Polymer flooding is mainly used as a mobility control agent and tries to minimize the effect of viscous fingering (Sorbie and Seright, 1992). Polymer flooding has a subtle impact on the permeability of the rock and some studies show that some polymers can change the permeability of the reservoir rocks to some extent by adsorption (Mishra et al., 2014).

On the other hand, gel treatment targets the permeability heterogeneity of the reservoir (Sorbie and Seright, 1992). The volume of the gel treatment often is a fraction of polymer flooding (Sorbie and Seright, 1992). There are wide ranges of conformance problems that can only be treated with gel treatment, and polymer flooding does not have any effect on them. For instance, water problems near the wellbore, conning from an aquifer and water breakthrough due to fracture or high permeability streaks are among issues that can only be treated with gel treatments (Sorbie and Seright, 1992).

Despite the difference between the functionality of polymer flooding and polymer gel treatments, some parameters are used commonly to measure the effectiveness of these technologies. In the following sub-sections, the most common parameters are introduced.

Mobility Ratio (M)

Mobility ratio, M, defined as the ratio of mobility of the displacing fluid (e.g., water) λ_w to the mobility of displaced fluid (e.g., Oil) λ_o (Imqam et al., 2015; Goudarzi et al., 2017; Imqam et al., 2018).M can be defined as:

$$M = \lambda_w/\lambda_o = (k_{rw}/\mu_w)/(k_{ro}/\mu_o) \quad (2.4)$$

where

λ_w= water mobility;

λ_o= oil mobility;

k_{rw}= relative permeability to water;

k_{ro}= relative permeability to oil;

μ_w= water viscosity;

μ_o= oil viscosity

Resistance Factor (F_r)

Resistance Factor, Fr, defined as the ratio of mobility of the water λ_w to the mobility of gelant/polymer ($\lambda_{gelant/polymer}$) (Imqam et al., 2015; Goudarzi et al., 2017; Imqam et al., 2018)

$$F_r=\lambda_w/\lambda_{gelant/polymer}= (k_{rw}/\mu_w)/ (k_{gelant/polymer}/\mu_{gelant/polymer}) \quad (2.5)$$

where

λ_w= water mobility;

$\lambda_{gelant/polymer}$= gelant or polymer mobility;

k_{rw}= relative permeability to water;

$k_{gelant/polymer}$= relative permeability to gelant/polymer;

μ_w= water viscosity;

$\mu_{gelant/polymer}$= gelant/polymer viscosity

Resistance factor (F_r) is useful for better understanding of the behavior of gel and/or polymers during injection. The F_r can also be expressed by the ratio of pressure drop for gel/polymer injection to pressure drop during water injection as follow:

$$F_r=\Delta P_{gel}/\Delta P_{water} \quad (2.6)$$

Equation (2.6) provides useful information about the injectivity of gel/polymers. Injectivity of the chemical is one of the important factors in designing the chemical conformance improvement technologies (Imqam et al., 2015; Goudarzi et al., 2017; Imqam et al., 2018).

Residual Resistance Factor (F_{rr})

Residual Resistance Factor, F_{rr}, defined as the ratio of mobility of the water or oil before and after gel treatment and/or polymer flooding (Imqam et al., 2015; Goudarzi et al., 2017; Imqam et al., 2018)

$F_{rrw} = (k_{rw}/\mu_w)_{Before} / (k_{rw}/\mu_w)_{After}$ (2.7)

$F_{rro} = (k_{ro}/\mu_o)_{Before} / (k_{ro}/\mu_o)_{After}$ (2.8)

where

k_{rw}= relative permeability to water;

k_{ro}= relative permeability to gelant/polymer;

μ_w= water viscosity;

μ_o= oil viscosity

Similar to the resistance factors, the residual resistance factors can also be expressed in term of pressure drop i.e., the ratio of pressure drop after gel treatment to pressure drop before gel treatment as follow:

$F_{rrw} = (\Delta P_w)_{After} / (\Delta P_w)_{Before}$ (2.9)

$F_{rro} = (\Delta P_O)_{After} / (\Delta P_O)_{Before}$ (2.10)

Adsorbed Layer Thickness (e)

The adsorbed layer thickness (e) is calculated from the relationship between pore throat size and residual resistance factor (Chauveteau et al., 2004). It can be estimated as:

$$e = rp(1 - Frr^{-1/4}) \quad (2.11)$$

where

e = thickness of adsorbed layer;

rp= pore throat radius;

F_{rr}= residual resistance factor.

3.2 Types of Gel Treatments

Gel treatments for conformance control practices are generally classified into three categories according to the type of the treated wells whether it is a producer or an injector, gel penetration depth, target problem and volume of chemical injected (Han et al., 2014). The technologies, respective applicable conditions, and their corresponding targeted problems are presented in Table 3-1 (Han et al., 2014). Table 3-1 also provides the advantages and disadvantages of each of the methods. Besides, knowing the terms used to describe gel treatments categories is essential for communication in the oil and gas industry.

Table 3-1: Types of gel treatment for conformance control (regenerated with permission from Han et al., 2014).

Treatment Types	Well Types	Treatment Diameter	Targeted Problems	Advantages	Disadvantages
Water Shutoff	Producer	3-30 ft	Thief zones, Water conning	Immediate Response	Low Success Rate and High Risky
Profile Modification	Injector	30-100 ft	High permeability zones	High Success Rate	Short-Lived Response
In-Depth Flow Diversion	Injector	0.1-.05 PV*	Cross-flow problems	Far-Wellbore Effects	Large Volume

*PV=Pore Volume

3.2.1 Water Shut-off (WSO)

As shown in Figure 3-2, water shutoff treatments are applied to the production well to correct the reservoir permeability heterogeneity near wellbore and to mitigate the early water breakthrough. Water shut off treatments can further be categorized as non-selective and selective treatments according to the permeability reduction level of the material used (Liu et al., 2010).

Non-selective water shut-off treatment. When there is an impermeable layer between oil and water zones as shown in Figure 3-2 (a), strong polymer gels should be applied to the high permeability water out zone to treat the water production problem near the wellbore. These types of treatment are called “non-selective water shutoff treatment.”

In these treatments, gels are in strong bulk form, block the high permeability water-bearing zones and divert the subsequent injected water into the low permeability zones (Liu et al., 2010).

Selective water shut-off treatments. When there is some level of vertical communication between high permeability and low permeability layers, there is a possibility of damage to oil bearing zone of the reservoir. Selective water shut off treatment with relative permeability modifiers is the suitable option to minimize the risk of the polymer gel treatment (Liu et al., 2010).Due to the danger of damage to oil-bearing zones by sealing materials, such as strong bulk gels, the gel placement techniques are essential for non-selective water shutoff treatments while selective water shutoff treatments by relative permeability modifiers (RPM) might be bullheaded without compromising the oil production (Sydansk and Seright, 2007).

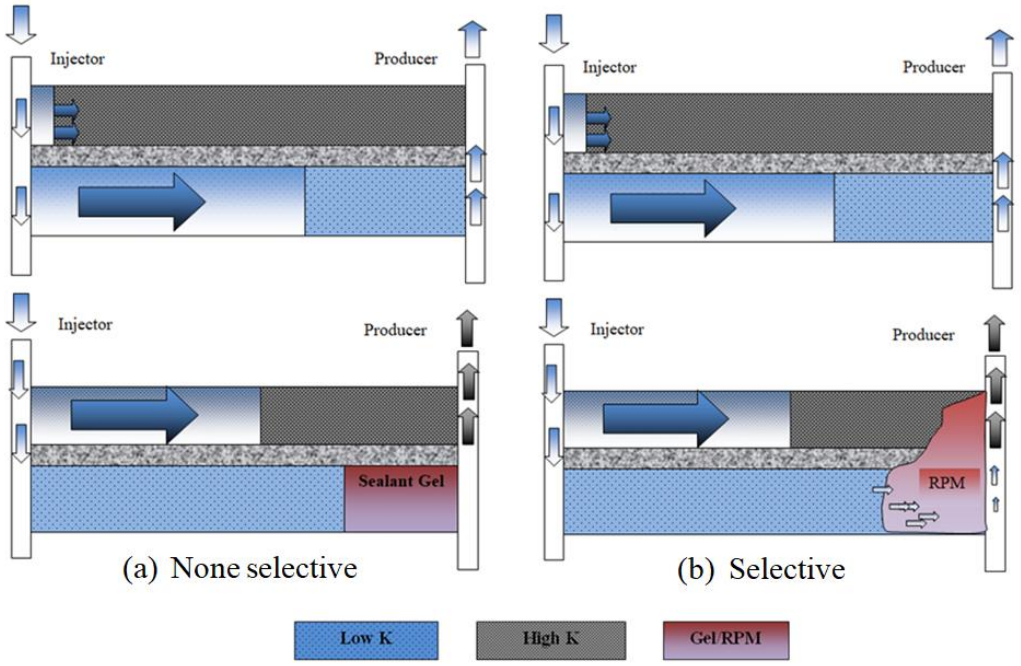


Figure 3-2: Water shutoff gel treatments methods, (a) non selective, and (b) selective.

An objective of a water shutoff treatment is the identification of the materials that can be injected into the production wells without mechanical zone isolation and that substantially reduces the water cut with minimum damage to oil-bearing zones (Sydansk and Seright, 2007). Mechanical zone isolation requires costly work over rigs operations

especially if the completion is gravel-pack or when the completion involved the subsea tieback flow-line (Seright, 2006b). Relative permeability modification (RPM) or sometimes called Disproportionate permeability reduction (DPR) is a property of some water-soluble polymers and polymer gels that can reduce the permeability of the porous media to water to more extent than oil (Liang et al., 2017). Therefore, selective water shut off treatments with bullhead injection of materials into all production layers are the most favorable water shutoff methods in oil and gas industry (Sydansk and Seright, 2007).

It is well-known that the selective water shut off treatments performance in field applications has varied between success and failure without understanding the exact reasons (Alfarge et al., 2018). Therefore, there is a necessity to understand the mechanisms that give this property to the polymer and gels, and the conditions that this type of treatment can be applied (Sydansk and Seright, 2007). Many researchers proposed different mechanisms for RPM behavior of the polymers and polymer gels (Liang and Seright, 1997; Liang and Seright, 2000; Liang et al., 1995; Zaitoun and Kohler, 1999; Alsharji e al., 1999; Alsharji et al., 2001; White et al., 1973; Willhite, 2002). The ten different RPM mechanisms proposed by different researchers are listed below:

- Wall effect /gel droplet mechanism (Liang and Seright,2000)
- Gravity effect mechanism (Liang et al., 1995)
- Lubrication/hydrophilic -film mechanism (Zaitoun and Kohler, 1999)
- Rock wettability change and water/oil pathways constriction (Zaitoun and Kohler, 1999)
- Capillary force and gel elasticity effect (Liang and Seright,1997)
- Polymer leaching from gel and reduction brine mobility mechanism (Liang and Seright., 1997)
- Gel swelling in water and shrinkage in oil (Alsharji e al., 1999)
- Polymer adsorption entanglement (Alsharji et al.,2001)
- Segregated pathway mechanism (White et al., 1973)
- Gel dehydration / deformation (Willhite, 2002)

Alfarge et al. (2017) reviewed, summarized and ranked the proposed mechanisms by researchers along with their weak points and opponents. This paper can be referred for

more details about relative permeability modification mechanisms. Sydansk and Seright (2007) provided some guidelines about when and where relative permeability treatments can be successfully applied.

3.2.2 Profile Modification

Sydansk & Romero (2011) defined "profile modification" as the mitigation and treatment of vertical conformance problems. However, in the field of petroleum engineering "profile modification" term widely used to differentiate the injection well treatment from production well treatment (Liu et al., 2006; Vossoughi, 2000; Yadav and Mahto, 2014). As shown in Figure 3-3 (a) injection fluids (water) can bypass low-permeability oil-bearing zones if the profile is not modified and cause low oil productivity and high water cut (Vasquez and Santin, 2015). Application of polymer gels in injection wells can reduce the permeability of the water-bearing zone and consequently improve the injection profile, Figure 3-3 (b).

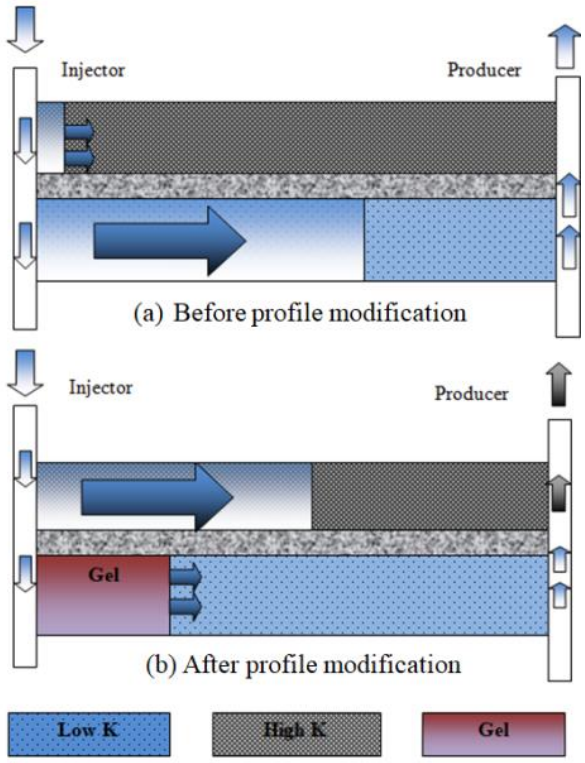


Figure 3-3: Profile modofication by polymer gels.

3.2.3 In Depth Flow Diversion (IFD)

This Technology was introduced in the late 1990s when the majority of oil fields have become mature with the low amount of oil near the wellbore, and many of the oil wells were already conventionally treated with plugging agents (Liu et al., 2010). When there is vertical pressure communication (cross-flow) between low permeability oil bearing and high permeability water-bearing zone or when gravity segregation is dominant, near-wellbore treatments are ineffective (Liu et al., 2010). This is because injected water returns to high permeability water out zone right after bypassing the placed treatments as shown in Figure 3-4 (a) (Liu et al., 2006). Therefore, to obtain more effective results, large volumes of treating materials are placed deep in the reservoir as shown in Figure 3-4 (b) (Liu et al., 2006). In-depth gel treatment has some advantages over near wellbore treatments especially when there is strong vertical pressure communication between layers. (Khames et al., 2017; Abdulbaki et al., 2014). IFD treatments are often sized to occupy about one-third of the distance between the injector and producer (Han et al., 2014).

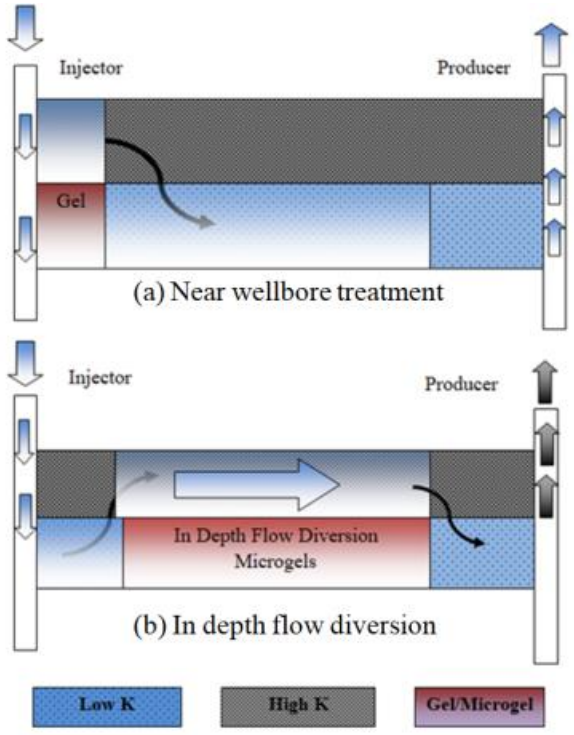


Figure 3-4: (a) Near wellbore treatment and (b) in-depth fluid diversion.

The differences between the objectives of water shut off treatment, profile modification and in-depth flow diversion, result in a necessity of selection of different chemical agents. For example, for non-selective water shut off treatment the ideal polymer gel system should have high sealing ability while for selective water shut off treatment, the relative permeability modification property is a key feature to be considered. These are mainly due to the risk associated to the water shut off treatment and possible damage to the production well. For profile modification application, the risk of damage to oil production does not exist while deeper gel penetration to the formation is required compared to water shut off treatments. Therefore, the selected polymer gel system should have adequate gelation time. In depth flow diversion application required a large volume of chemicals to be placed deep into the formation. In this case, the economics of the treatment, gelation time, in deep permeability reduction and long term stability of the polymer gel systems need to be considered pre-treatment. The next section of this paper devoted to the review of most widely applied polymer gel systems. The properties of gel systems such as gelation time, gel mechanical strength, selectivity, relative permeability modification, and sealing ability are addressed based on both field applications and lab experiments.

CHAPTER 4

Polymer Gel Systems

This chapter reviews the literature on the six commercially available polymer gel technologies in conformance control applications. Polymer gel systems were categorized into two main groups as conventional in-situ bulk gels and novel microgels. For each of the six selected technologies both field application results and relevant laboratory experiments are reviewed. The introduction to the technologies, their development, effect of reservoir condition (temperature, salinity, pH, etc.) and other important information related to the performance of these technologies are explained in detail. The technologies are also compared based on their properties, advantages, disadvantages and reservoir conditions.

4.1 Conventional In-Situ Bulk Gels

Conventional in-situ bulk gels are the most widely applied polymer gels in the field of conformance control (Brattekas et al., 2016; Salimi et al., 2014; Vasquez et al., 2008). These types of gel systems are prepared by a cross-linking of polymers with cross-linkers (Moradi, 2000). For the implementation of this type of technology, a gelant (solution of polymers+ cross-linkers+ water) is prepared in the surface and injected into the reservoir (Bai et al., 2015). Later with time and effect of reservoir temperature, the cross-linking reactions begin and turn the flow-able gelant into the bulk gel (Sydansk, 1990). The typical in-situ bulk gel consists of 5000-10000 ppm polymer, 500-2000 ppm cross-linker, and the remainder of the gelant solution being water (above 98%) (Moradi, 2000). Various types of polymers and cross-linkers have been used to prepare bulk gels. Polymers such as synthetic polyacrylamide polymers and biopolymers such as Xanthan gum are the most widely applied polymers in the upstream oil industry (Bai et al., 2015). Polyacrylamide polymers, including non-hydrolyzed polyacrylamide (PAM), partially hydrolyzed polyacrylamide (HPAM) and polyacrylamide/tert-butyl acrylate (PAtBA) are the most widely used types of polymer in conformance control (Ghriga et al., 2019). The cross-linkers mainly categorized into two types:

Metallic cross-linkers these are a type of cross-linkers that are making ionic bonding with polymers to form bulk gels. These are multivalent cations such as Cr (III),

Cr (VI) and Al (III) cations attached to some ligands such as malonate, lactate, citrate, propionate, and acetate. The ligand in the cross-linked structure is mainly used to control the rate of cross-linking reaction between metallic cross-linkers and polymer chains (Sydansk, 1990; Seright et al., 2006; Morgan et al., 1997).

Organic cross-linkers these are types of cross-linkers that are making covalent bonding with polymers. Various kinds of organic cross-linkers from formaldehyde source (e.g., hexamethylenetetramine (HMTA)), phenol source (e.g., hydroquinone (HQ)) and more environmental friendly such as polyethyleneimine (PEI) have been reported in the literature. Organic cross-linkers, due to the strong covalent bonding, are generally making stronger bulk gels compare to metallic cross-linkers, and they can resist higher temperature (Bai et al., 2015; El-Kasrani et al., 2014b).

From the different types of conventional bulk gel systems, HPAM/Cr(III) acetate and PAtBA/PEI are more widely used in oil fields (Alshammari et al., 2018; Fakher and Bai, 2018; Liang et al., 2017; Beltagy et al., 2015). This might be mainly due to their availability, lower cost, being less toxic and successful feedbacks from actual field implementations. Similar to other conventional in-situ bulk gels which are formed in the reservoir condition, the critical parameters such as gelation time, gel strength, selective penetration, depth of penetration and gel stability of these systems can significantly be affected during and after the injection into the reservoir. The properties, field application results, development, and other relevant topics related to these two widely used polymer gel systems are provided in the following sections.

4.1.1 HPAM/Cr (III) Acetate

In 1984, Sydansk in Marathon Oil Company patented HPAM/Cr (III) acetate gel system for conformance control applications (Sydansk, 1990). HPAM/Cr (III) acetate gel technology consists of forming aqueous gels by cross-linking partly hydrolyzed polyacrylamide polymers with Cr (III)-carboxylate groups (Sydansk, 1990). The cross-linking agent consists of Cr (III) ions and acetate (low molecular weight carboxylate anion). The polymer and cross-linker are attached through ionic bonding (Sydansk et al., 1990). Chromium (III) acetate is the preferred cross-linking agent because it provides overall longer gelation time, stronger gel and extended stability at reservoir condition

compared to other Cr (III) compounds and it has the advantage of being less toxic than Cr (VI) based technologies (Sydansk, 1990; Vargas and Zeron, 2008).

Since its development, this system successfully implemented in both sandstone and carbonate reservoirs throughout the world (Fakher and Bai, 2018). From 1989 through 1992, the Big Horn Basin of Wyoming alone had incremental oil recovery of 1 200 000 barrels of oil due to treatment of 17 injectors and 18 producers by HPAM/Cr (III) acetate gel system (Seright and Liang, 1994). More than 98% of the total incremental oil recovery attributed to injector's treatment, while the producer's treatment resulted in a reduction of water cut along with a decrease in oil flow rate (Seright and Liang, 1994). At low permeability contrasts between water-bearing an oil-bearing zone, the gelant tends to invade both oil and water layers (Seright and Liang, 1994). In one of the treatments mentioned above, the volume of the HPAM/Cr (III) acetate gelant injected was ten times more than the predicted volume of the thief zones. Further core and well test data demonstrated that the few successful water shut-off treatments at Big Horne Basin conducted in producers that intersected fractures with aquifers. While unsuccessful water shut-off treatments were in either matrix problems or the fractures that intersected both oil and water zones (Seright and Liang, 1994).

In the case of injector treatments, the near wellbore geology is less critical, because the possible damage to oil permeability occurs far away from the producers. The gel treatment implemented at Guarda oil field in Colombia in 2008 (Moreno et al., 2014) is a new successful field application of HPAM/Cr (III) acetate gel system for injection profile treatments. To correct injection profile of one of the injector wells at Guarda oil field, 25,736 bbls of HPAM/Cr (III) acetate gelant at the polymer concentration ranged from 2,000 to 8,000 ppm injected without zonal isolation. As Figure 4-1 shows, the oil rate increased post-treatment while rate of water production decreased (Moreno et al., 2014).

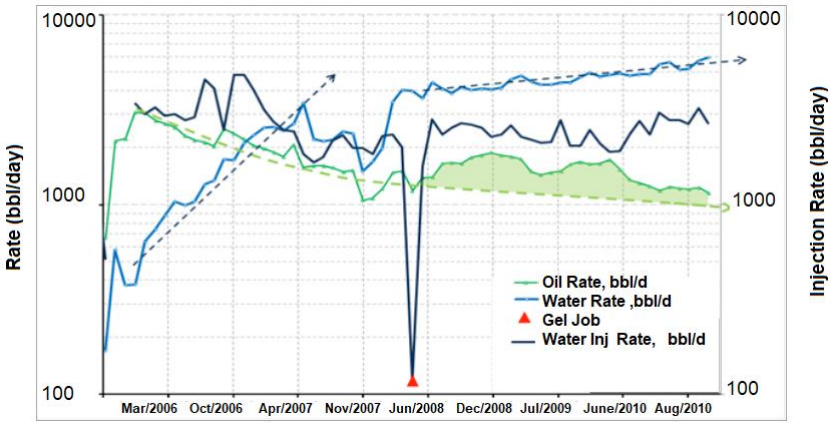


Figure 4-1: Pre and post-treatment production (reprinted with permission from Moreno et al., 2014).

During April-August of 2009 at Big Horn Basin, seven producers treated with 11,400 bbls of HPAM/Cr (III) acetate solution at polymer concentration ranged from 3,000-10,000 ppm. Despite the zonal isolation, the oil-bearing zone was damaged and resulted in a decrease in oil rate production by 56% (Bybee, 2011).

The goal of gel treatment technology is that of recognizing chemicals with RPM properties that can be bullheaded into any production well without significantly impairing oil productivity (Han et al., 2014). The extent to which HPAM/Cr(III) acetate bulk gel can provide RPM effect depends on factors such as permeability contrast between water and oil zone, the leak-off distance of the gel, gel system composition, types of conformance problem (Linear or Radial) and placement strategies (Seright, 2009). Seright (2009) studied the DPR effect of pore-filling chromium (III)-acetate- hydrolyzed polyacrylamide system in both radial and linear systems. He demonstrated that some systems of HPAM/Cr(III) acetate pore-filling gel can have final residual resistance factor of greater than 2000 for water and as low as 2 for oil. As Figure 4-2 shows, for fracture problems without cross flow, one-foot gel leak-off into the adjacent matrix in water and oil-bearing zone can provide different residual resistance factors to oil and water. F_{rrw} of 1000 means that the water should pass through an equivalent 1000 ft of untreated matrix rock to be produced into the fracture while F_{rro} of 10 means that oil should pass through only 10 ft of the equivalent of untreated rock matrix to produce into the fracture. In the treatment of linear

systems, there is no need to achieve specific low oil residual resistance factor and only that the water residual resistance factor be higher is reliable (Seright, 2009).

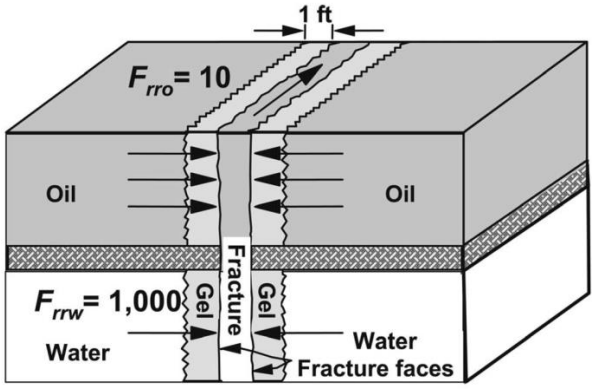


Figure 4-2: The application of pore-filling Gel to restrict water entry into the Fracture (reprinted with permission from Seright, 2009).

As Figure 4-3 shows, a gelant solution containing 5,000 ppm of HPAM and 417 ppm of Cr^{3+} provided residual resistance factors of 700 and 4.8 for water and oil, respectively. Moreover, the author concluded that for matrix problems (radial low), the oil residual resistance factor of more than 2 might be considered unacceptable (Seright, 2009).

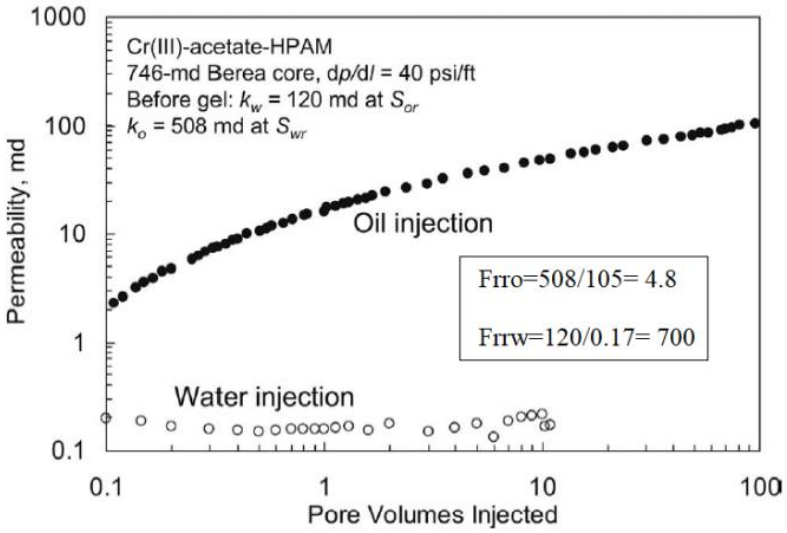


Figure 4-3: Water and oil permeabilities after HPAM/Cr (III) acetate placement in Berea core (reprinted with permission from Seright, 2009).

Fakher and Bai (2018) used data from more than 1050 experiments to perform data analysis and to provide screening criteria for HPAM/Cr (III) acetate gel system. A

mathematical model was generated and validated in part of their work. As Figure 4-4 shows, as the polymer concentration increases, the residual resistance factor to oil also increases; the increase is exponential, which shows that even slight change in polymer concentration would cause in a significant change in oil residual resistance factor. The correlation obtained from their work had a high R^2 value and the data used for validation was not included in the generation of the equation. Equation 4.1 as follow:

$$F_{rro}=0.0353e^{0.0018p} \quad (4.1)$$

where F_{rro} is the residual resistance factor to oil, e is Euler's Number, and p is polymer concentration in ppm.

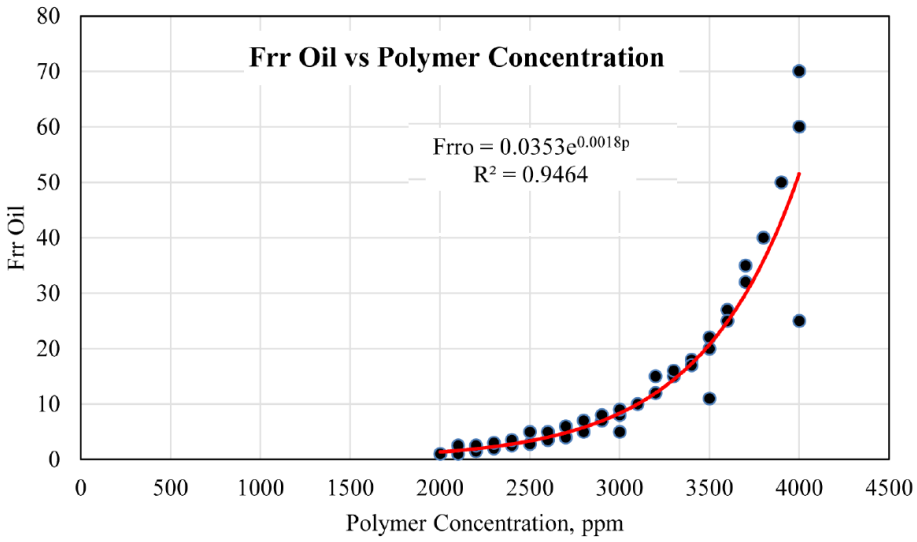


Figure 4-4: F_{rro} vs. polymer concentration (reprinted with permission from Fakher and Bai, 2018).

Seright et al. (2006) used X-ray computed micro-tomography (XMT) to study the RPM mechanisms of pore-filling Cr (III)-acetate-HPAM gel system. They demonstrated that residual resistance factor to water was high because water passed through the gel structure while for oil, dehydration of gel and forced channeling of the oil provided smaller residual resistance factor to oil. Gel dehydration/deformation by oil and segregated pathways for oil and water as the RPM mechanisms were later studied using both nuclear magnetic resonance (NMR) and micro-glass-models (Laing et al., 2017).

Figure 4-5 shows a summary of the results of the sub-micro-glass model. First, the oil extruded through the pore-filled gel and caused channels to open to oil. At this stage, gel dehydration and shrinkage was the primary mechanism. Second, the water passed through the channels that reopened to oil flow. Later, the gels started to rehydrate and narrowed the channels. At this stage, residual oil saturation further restricted the channels flow capacity to water. Finally, further water flooding caused the channels to close, and water started passing through the gel body (Liang et al., 2017).

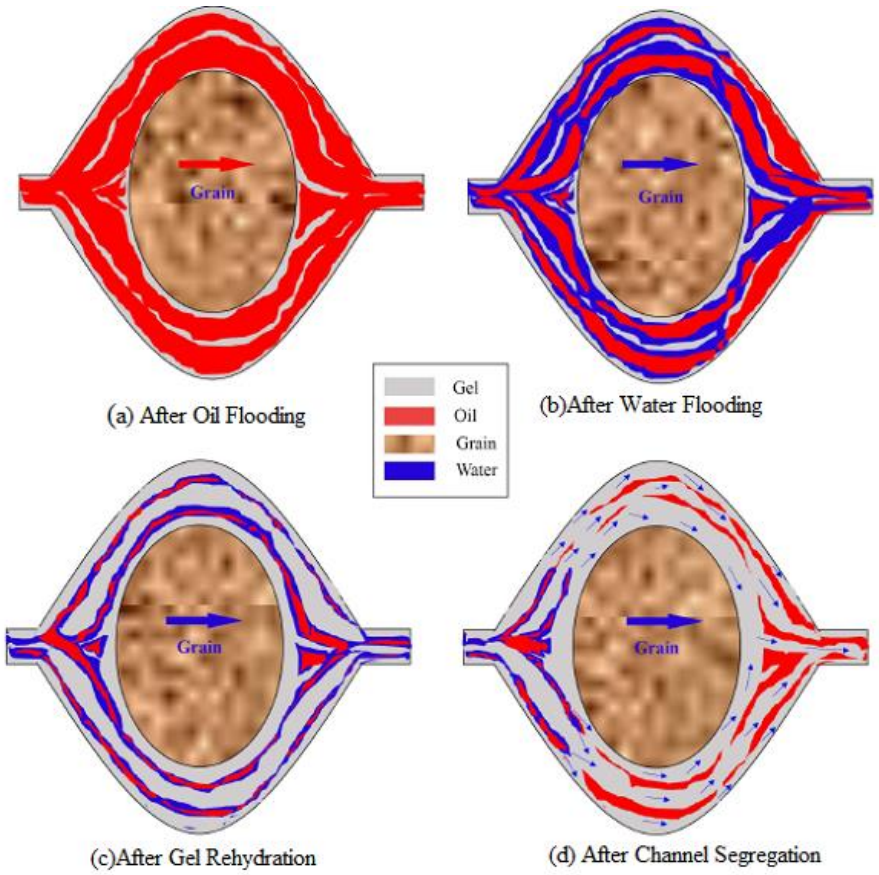


Figure 4-5: Schematic diagram of the mechanisms causing DPR (reprinted with permission from Liang et al., 2017).

Since HPAM/Cr (III) acetate gelant is composed of low and high molecular weight components, chromatographic separation, precipitation, and diffusion may cause uneven distribution of Cr^{3+} and HPAM near wellbore and affect gelation (Pu et al., 2018 a). Ganguly et al. (2001) observed that HPAM concentration inside the fracture did not change during 17 hours of shut-in time, while Cr^{3+} concentrations decreased from 100 to 20 ppm

due to the diffusion of chromium ions to the Berea core matrix. They further mentioned that the pH augmentation by the dissolution of carbonates might cause chromium ion precipitation (Ganguly et al., 2001). Pu et al. (2018a) studied the effect of Cr^{3+} diffusion on the gelation of HPAM/Cr (III) acetate system through a dialysis bag method. They demonstrated that Cr^{3+} diffusion affects the gelation of HPAM/Cr^{+3} acetate system with varying the initial HPAM polymer concentration, HPAM molecular weight, the Cr^{3+} initial concentration and degree of initial HPAM hydrolysis. They determined that the Cr^{3+} concentrations in the initial gelant and final gel system are different, and neglecting the diffusion effects can lead to overestimating the gelation in field applications (Pu et al., 2018a).

Polymer concentration, degree of hydrolysis of polymer, polymer molecular weight, cross-linker concentration, polymer to cross-linker ratio, divalent cations, temperature, salinity, pH and shear are factors that are controlling the reliability of HPAM/Cr (III) acetate gel systems in conformance control applications. These factors control the gelation time, gel syneresis, gel strength and stability which consequently affect the effectiveness of the gel system for near wellbore water shut off treatments and ability to divert flow deep into the reservoir (Sydansk, 1990; McCool et al., 2007; Karimi et al., 2016; Wang et al., 2016).

Karimi et al. (2016) studied the effects of different parameters such as polymer concentration, cross-linker concentration, polymer to cross-linker ratio, pH, and temperature on the syneresis of HPAM (15%) hydrolyzed –Cr (III) acetate gel system. They demonstrated that, by increasing polymer concentration and cross-linker concentration, the percentage of syneresis decreases and increases, respectively. The best range of pH to reduce the onset of syneresis was between 5.5 and 7.5. The results showed that, after six months, no syneresis was observed for temperatures below 60 °C, while increasing the temperature from 80 °C to 100 °C reduced the onset of syneresis starts time from more than 20 days to less than four days. Vargas and Zerón (2008), also reported the problem of thermal hydrolysis and syneresis of HPAM/Cr (III) acetate gel system at reservoir temperature above 60°C and Sydansk (1990) suggested the use of optimum cross-linker concentration to prevent the early syneresis of this gel systems. They further find out

that, to achieve the maximum stability at the temperature of 80°C, the optimum polymer to the cross-linker ratio is 40 to 1 (P/C=40) (Karimi et al., 2016). Shear also induces the syneresis of HPAM Cr (III) acetate gel system and gel systems exposed to higher shear stress experienced more syneresis (McCool et al., 2007).

Wang et al. (2016) conducted a laboratory study on the effect of different parameters on gelation time, long term stability, and gel strength and oil recovery improvement of HPAM/Cr (III) acetate gel system. Figure 4.6 summarizes the results of their experiment on gelation time determination on gel systems composed of same Cr (III) concentration of 100 mg/l and various polymer concentrations at 60 °C, 80 °C and 95 °C. It has been shown that gelation time decreases with polymer concentration increases. Temperature is the most critical factor controlling the gelation time of HPAM/Cr (III) acetate gel systems and limits the ability of the gel to penetrate deep into the reservoir. For example, as shown in Figure 4.6, the gelation time of the gel system with a polymer concentration of 4000mg/l reduced from 6.0 hours at 60 °C to 45 minutes at 95 °C which is too short of providing in-depth gel treatment (Wang et al., 2016).

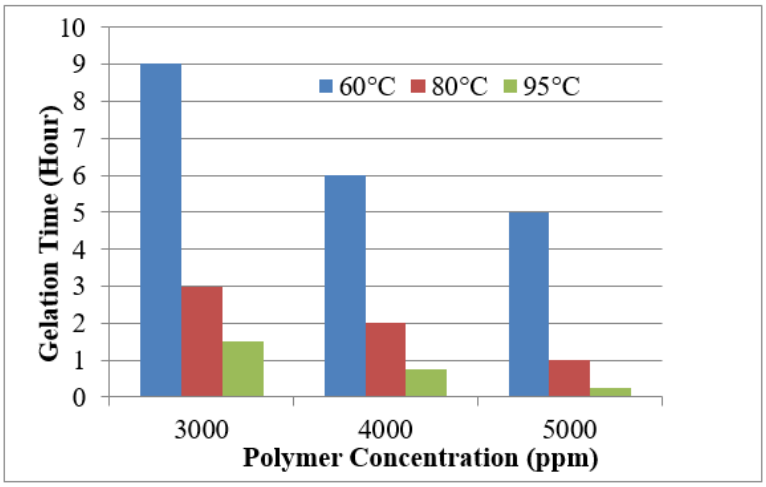


Figure 4-6: Gelation time by bottle test for gelants with different polymer concentration and 100mg/l of Cr (III) (data points visually selected from Figure 3 in Wang et al., 2016).

Maturity of the gel system, salinity of the formation water and salinity of makeup brine are the other vital factors for the application of HPAM/Cr (III) acetate gel systems, especially for fractured carbonates reservoirs. Brattekas et al. (2015) determined that mature gel can provide higher residual resistance factor to water compared to immature gelant. They demonstrated that after the injection of 120 PV of water, substantially higher residual resistance factors to water were observed in cores treated with mature gels (F_{rrw}=5,000) than the core treated with immature gelant (F_{rrw}=600).

Salinity and hardness (divalent cations) are also limiting factors for the application of HPAM/Cr (III) acetate gel system in water shut off applications. For example, 500 mg/L of hardness (Ca^{2+}, Mg^{2+}) at a temperature of 88°C can lead to fast precipitation of HPAM polymers (Moradi, 2000). The maximum salinity for this system reported as 30,000 ppm (Moradi, 2000). The difference between the salinities of brine and the water come in contact with gel in the reservoir influences the plugging efficiency of the HPAM/Cr (III) acetate gel system. Brattekas et al. (2016) proved that low-salinity chase waterfloods could improve the blocking capacity of the mature gel in fractures.

Since the harsh conditions (high shear rate, temperature, and high salinity) of oil reservoirs lead to gel degradation, it seems necessary to improve the gel thermal, chemical and physical stability (Cordova et al., 2008; Johnson et al., 2010; Singh et al., 2018). In recent years, there has been an increasing interest in the application of nanocomposite in HPAM/Cr(III) acetate gel structure (Cordova et al., 2008; Johnson et la., 2010; Salimi et al., 2014; Singh et al., 2018; Asadizadeh et al., 2018). Salami et al. (2014) investigated the effects of clay on various properties such as thermal strength and elastic properties of the nanocomposite gel system. They demonstrated that the addition of montmorillonite to the HPAM/Cr (III) acetate gel system leads to delay in thermal degradation and improvement of its elastic properties. Singh et al. (2018) also found that a nanocomposite gel containing nano fly ash has better gelation strength, gelation time and plugging efficiency than non-nanocomposite gel.

To delay the gelation time of HPAM/Cr (III) system and reduce the loss of cross-linker by chromatographic separation, polyelectrolyte complex (PECs) system consists of polyethylenimine (PEI) and dextran sulfate (DS) were used to sequester Cr (III). The cross-

linker is hide from the polymer in a nano-composite complex and thereby the gelation time of the HPAM/ Cr (III) system can be delayed (Cordova et al., 2008; Johnson et al., 2010). Johnson et al. (2010) demonstrated that the encapsulation of chromium in PECs complex can delay the gelation time of a HPAM/Cr (III) system (0.5 % HPAM + 0.01%Cr (III) from less than 30 minutes to seven days at 40°C. However, application of this technology at higher temperature is not reported in the literature. Asadizadeh et al. (2018) evaluated the application of a new nano-composite polymer gel system. They introduced the addition of SiO_2 nanoparticles to HPAM/Cr (III) acetate gel system. To evaluate the effect of SiO_2 on gel strength and thermal stability of HPAM/Cr (III) acetate technology at various salinities, they performed some set of experiments. Table 4-1, summarized the results of their experiments. The polymer gel systems all cured at 100°C. As shown in Table 4-1, addition of 2000 ppm SiO_2 nano-particles to HPAM/Cr (III) acetate systems increased the gel strength (elastic modulus) and gel thermal stability (inflexion temperature) of the gel systems prepared in sea and formation waters. However, effect of this new nanocomposite on other properties of the gel such as, propagation in porous media and relative permeability modification need to be further study.

Table 4-1: Effect of SiO_2 nano-composite on gel strength and thermal stability of HPAM/Cr (III) acetate at high and low brine salinity (data collected from experiments in Asadizadeh et al., 2018).

Polymer gel composition	**Gel strength**	**Thermal Stability**
10000 ppm HPAM+ 10 P/C %wt+ Sea water	12.5 Pa	140.8 °C
10000 ppm HPAM+ 10 P/C %wt+ Sea water+2000 ppm SiO_2	13.56 Pa	157.9°C
10000ppm HPAM+ 50 P/C %wt+ Formation water	9.8 Pa	135.2°C
10000 ppm HPAM+ 50 P/C %wt+ Formation water+2000 ppm SiO_2	11.57 Pa	145.7°C

4.1.2 PAtBA/PEI

In 1997, Morgan and co-workers introduced a unique polymer gel system based on the cross-linking of polyacrylamide/tert-butyl acrylate (PAtBA) and polyethylenimine (PEI) (Morgan et al., 1997). The introduction of this system was to overcome the problems

related to HPAM/Cr (III) acetate gel systems such as precipitation of the cross-linker in carbonate rocks, low thermal stability and gelation time at high reservoir temperature (Morgan et al., 1997). The first solution to solve the low gelation time relied on delaying cross-linking between cross-linker and the negatively charged polymer by controlling the degree of hydrolysis of the polymer (Hardy et al., 1999). PAtBA copolymer was offered based on its controllable hydrolysis, high solubility in water and feedstock price (Morgan et al., 1997). The problems associated with cross-linker itself, such as precipitation in carbonate rocks (at high pH) and toxicity encouraged the investigators to find a new cross-linkers. PEI was selected due to its eco-friendly aspects and ability to form covalent bonds with PAtBA to form a firm gel (Morgan et al., 1997). The covalent bonding for PAtBA/PEI system is a stronger bonding compared to ionic bonding between Cr (III) and HPAM. The initial experiments showed that this gel system could withstand a temperature of 156 °C for two months in bulk form (Morgan et al., 1997). In 1998, Hardy et al. (1999) found that this gel system has both excellent thermal stability and propagation properties in porous media; therefore, they commercialized it (Hardy et al., 1999).

The gelation time, gel strength and propagation of PAtBA/PEI gel system have been the subjects of many studies during the last two decades. The effect of parameters such as temperature, pH, polymer concentration, cross-linker concentration, mixing water, inorganic salts, retarders, contamination with ferric iron, and addition of solid particles on gel performance are investigated.

Al-Muntasheri et al. (2007) studied the effect of different parameters on the gelation time and gel stability of this system. The summary of the results as follows:

Mixing water effect: The gelation time of gel prepared in seawater was double the gelation time of gel prepared in distilled water.

Monovalent cations effect: Sodium ions and potassium ions (Na^+, K^+) both delayed gelation time and effect of the monovalent ion with higher charge density (ionic charge/size) was more pronounced.

Divalent cations effect: Ca^{2+} cations increased gelation time and effect was more than monovalent cations due to higher charge density.

Initial pH effect: Acidic pH decreased the gelation time and adversely affected the gel stability. A pH of at least 8 required for a stable gel to form.

Temperature effect: Gelation time decreased as temperature increased.

Polymer concentration effect: Gelation time decreased as polymer concentration increased.

Cross-linker concentration effect: Gelation time decreased as cross-linker concentration increased.

Ferric iron concentration effect: High concentration (1000 mg/L) reduced gelation time and gel stability (to less than a few hours).

Gelation times reported for PAtBA/PEI gel system with polymer loading of 3 to 9 wt% and in the temperature range of 70 to 150° C varied from 0.3 to 15 hours (Deolarte et al., 2009). PAtBA/PEI gel system with thermal stability at a temperature of 191 °C reported in the literature (Deolarte et al., 2009).

Measurement of gel strength showed that the PAtBA/PEI formed remarkably stronger gel compared to HPAM/Cr (III) acetate (Al-Muntasheri et al., 2007). Liu and Seright, (2000) demonstrated that a typical PAtBA/PEI gel system (Prepared with 7wt% PAtBA and 0.3 wt% PEI) cured at 150°C for 12 hours, has an elastic modulus of 700 Pa while a typical HPAM/Cr (III) acetate gel system (Prepared with 0.5wt% HPAM and 0.0417wt% Cr^{3+}) cured at 41°C for 24 hours have an elastic modulus of only 7 Pa.

The performance in porous media was also studied. PAtBA/PEI gel in porous media had good injectivity and showed to be eight times faster than HPAM/Cr (III) acetate gel system under similar conditions (Bai et al., 2015). Vasquez et al. (2005) studied the permeability reduction of this gel system in Oklahoma sandpacks. They demonstrated that a permeability reduction of up to 88% was achieved and maintained at a high temperature of 176.6 °C for an extended period.

All these good results in both bulk tests and core flood experiments encouraged the operators to use PAtBA/PEI in water shut-off field applications. Despite the variety of gel

strength, gel stability and gelation time than can be obtained by traditional PAtBA/PEI system, several additional techniques and materials were used in both lab experiments and field applications to enhance the performance of this gel system further.

In the rest of this section, these techniques along with the results of their field or lab observations discussed.

Use of chemical retarders to delay gelation time. Various methods are used to delay the gelation time of PAtBA/PEI gel system for water-shut off application. These methods include varying polymer and cross-linker concentrations, adjusting the pH and cooling the near-wellbore area with water pre-flush (Al-Muntasheri et al., 2010). However, the use of inorganic salts such as sodium carbonate (Na_2CO_3), ammonium chloride (NH_4CL) and sodium chloride (NaCl) as retarders was the most economical method to delay the gelation time of PAtBA/PEI gel system.

Sodium carbonate (Na_2CO_3) reported increasing the gelation time of a PAtBA/PEI gel system from 1h at 126°C to 6 h at 176.6°C (Vasquez et al., 2005). The mechanism for delaying the gelation by this retarder is believed to be an interaction between the sodium cations of the retarder and carboxylate groups of the polymer, thus occupying the cross-linking sites and consequently delaying the gelation time. Eoff et al. (2007) after doing a sandpack flow test using this retarder at 176.6°C concluded that the sodium carbonate (Na_2CO_3) did not adversely affect the gel strength and gel propagation properties while increased the working temperature of the gel system.

Sodium carbonate (Na_2CO_3) showed some disadvantages when used in a horizontal well with the high salinity and reservoir temperature of 149°C in Saudi Arabia (Al-Muntasheri et al., 2010). They found that the sodium carbonate was incompatible with mixing brines and formation brine of the subject field (white precipitate observed in gelling solution). Al-Muntasheri et al. (2010) substituted sodium carbonate with sodium chloride. Sodium chloride (NaCl), on the other hand, did not have compatibility with brine and formation water but its retardation effect did not meet the field expectations. Thus, it drove further research to find a new retarder that was compatible with mixing brine, cost-effective and efficient. Ammonium chloride (NH_4Cl) was reported to succeed in delaying the

gelation time of the traditional PAtBA/PEI gel system at 150 °C to 90 minutes without compatibility problems. The treatment with this new retarder showed 46% reduction in water cut and 17 times more hydrocarbon production in one of the gas wells of a field in Saudi Arabia. Despite these results, further research found that NH_4Cl addition into the PAtBA/PEI gel system results in weaker gels compared to salt-free gel systems or when NaCl used in the mixing brine (El-Kasrani et al., 2014a). As we mentioned earlier, the mechanism for these retarders to delay gelation time believed to be the effect of positive ions such as Na^+ on carboxylate groups of the polymer. In the following subsection, another method for gelation time elongation which affect the cross-linker (PEI) rather than the polymer (PAtBA) reported. Table 4-2 summarized the different chemical retarders used to delay the gelation time of PAtBA/PEI gel system. The maximum temperature, effect of retarders on final gel strength and their compatibility with high brine salinity of the reservoirs are also compared. From the information discussed, there is a need to find or develop new effective retarders that do no reduce the gel strength.

Table 4-2: Summary of chemical retarders used for PAtBA/PEI gel systems and their characteristics (data collected from Vasquez et al., 2005; Al-Muntasheri et al., 2010; Eoff et al., 2007).

Retarders	Temperature	Gel strength	Brine salinity
Sodium Carbonate (Na_2CO_3)	177°C	Strong gel	Incompatible with brine
Sodium Chloride (NaCl)	126°C	Strong gel	Compatible with brine
Ammonium Chloride (NH_4Cl)	150°C	Weak gel	Compatible with brine

Use of chemical derivatives to delay gelation time. Another method to delay the onset of cross-linking of traditional PAtBA/PEI gel system reported in the literature is to alter the PEI chemistry to reduce its activity. Hardy et al. (1999) found that chelating of polyethylenimine with zirconium can increase the gelation time of a classical PAtBA/PEI by a factor of two at 100 °C. Polyamino acid was also observed to form a complex with polyethylenimine (PEI) and delay the cross-linking reaction (Vasquez et al., 2006). In the mentioned systems the amine groups of the PEI hide from the polymer and cause gelation

delay. Another system was derivatized-PEI (d-PEI), where amine groups of the PEI were converted to amides to delay gelation. The PAtBA/d-PEI gel system provided a gelation time of 13 h at 149°C compared to 0.3 h at 130 °C for traditional PAtBA/PEI gel system. The PAtBA/d-PEI also showed 100% permeability reduction at the temperature of up to 190 °C when used in dynamic sandpack flow experiments (Vasquez et al., 2006).

Use of solid particles for gel-strength enhancement. To enhance the strength of PAtBA/PEI gel systems, other materials such as cement, silica flour and rigid setting materials added to this system. Van Eijden et al. (2004) studied the effect of cement in the PAtBA/PEI gel system at two producer wells in Syrian oil fields. The initial lab results showed that the modified PAtBA/PEI gel system withstood different pressures up to 180 bars and had excellent sealing behavior in sandpacks. These results encouraged the operators to apply the new system in the field. In the first well, with the bottom-hole temperature of 118°C, modified gel system increased the oil rate from 3,000 BOPD to 4,000 BOPD, and water cut decreased from the initial value of 63% to 25%. However, throughout one-year water cut increased from 25% to 55%. The second well treatment, with a bottom-hole temperature of 144°C, was not successful.

PAtBA/PEI/cement system showed some drawbacks such as the difficulty of treatment design and interaction between cement and retarders used to elongate gelation time. These drawbacks encouraged to substitute cement by other inert materials. Silica flour was therefore selected to replace cement due to its cost-effectiveness, availability and more importantly its inert nature (Van Eijden et al., 2005). Vasquez et al. (2008) also confirmed the inert nature of silica flour through gelation-time measurements of PAtBA/PEI/retarder gel systems with and without silica flour. Figure 4.7 illustrates the gelation times for both neat samples (PAtBA/PEI) and the filtrate (PAtBA/PEI/silica-flour). The proximity of the blue and red lines in the Figure 4.7 suggested that the existence of silica flour in the gel system does not impact the gelation time of the system.

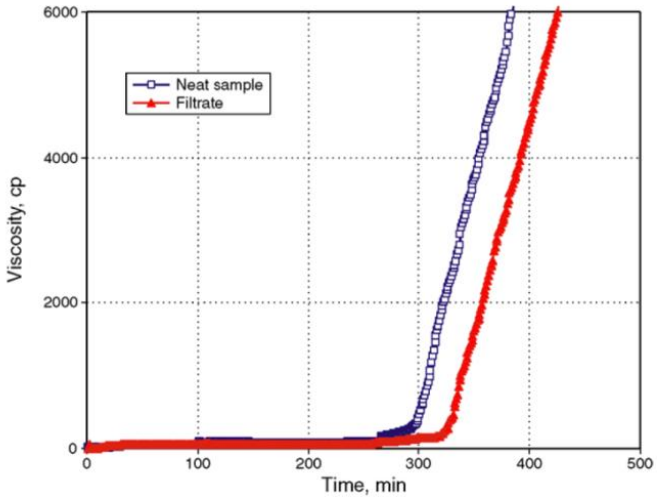


Figure 4-7: Gel times for neat sealant and filtrate (reprinted with permission from Vasquez et al., 2008).

The effect of silica flour percentage on the sealing ability and leak-off percentage of the gel system thus investigated (Van Eijden et al., 2005). The results showed that high silica flour loading (above 50 wt %) was required to effectively seal the porous media (Van Eijden et al., 2005). Moreover, the modified PAtBA/PEI/silica flour showed to be able to withstand pressure gradient up to 172 bars (Van Eijden et al., 2005). The new gel system successfully, therefore, employed in total sealing of a long perforation interval (186 m) of a well with the bottom-hole temperature of 148°C in the Syrian oil field. The main drawback of the PAtBA/PEI/Silica-flour gel system was its high silica-flour loading (50 wt %) requirement (Van Eijden et al., 2005).

Most recently, Beltagy et al. (2015) reported the use of both PAtBA/PEI/retarder and PAtBA/PEI/retarder/silica-flour gel systems in a high-temperature (160.5 °C) well in the Saqqara field in the Gulf of Suez, Egypt. The mentioned well was producing with water-cut off 95%, after only one year of production. The early water production was due to the early water breakthrough through one of the productive zones (zone number 3 among four productive zones). A PAtBA/PEI/retarder gel system was used to seal the troublesome zone while PAtBA/PEI/retarder/silica-flour (50 wt %) used for temporary isolation of other zones. The conformance treatment was successful and decreased water production from 2300 BWPD to almost zero BWPD (Beltagy et al., 2015).

Deolarte et al. (2009) described the application of another solid particle additive referred to as rigid setting materials (RSM). On the contrary to the silica flour, rigid setting materials (RSM) required in lower loading percentages (<5 wt %). This system was first introduced to treat the near-wellbore water production problem in a well at the Cantarell oil field in Mexico. The PAtBA/PEI was used initially to treat the matrix problem. Since the PAtBA/PEI gel system formed relatively deep from the wellbore, the very near wellbore area left untreated, and water finds its way into the wellbore after passed the treated layer. This problem observed until PAtBA/PEI/RSM gel system introduced to the wellbore. The RSM is metal oxy-chloride type cement that reported to have thermal stability up to 204 °C and to develop high compressive strength of up to 275 bars in a few hours. Since the modified PAtBA/PEI/RSM system formed in a short period, it adequately sealed the near-wellbore challenging problem. This new technology was later applied in other wells in the Cantarell oil field in Mexico, and in some cases, the zero water cut observed from the treatment (Deolarte et al., 2009).

Currently, more than 1,000 PAtBA/PEI system treatments have been performed globally to address different conformance problems, including zonal isolation, casing integrity issues, wellbore integrity, fracture shut-off, high-permeability streaks, and water coning/cresting (Vasquez and Santin, 2015; Alshammari et al., 2018).

4.2 Novel Microgels

The drawbacks related to conventional in situ bulk gels such as lack of gelation control at high reservoir temperature, chromatographic separation, and difficulty to provide in-depth flow diversion encouraged the researchers to developed novel polymer gel systems (Mack and Smith, 1994; Coste et al., 2000; Chauveteau et al., 2004; Pritchett et al., 2003). The most widely applied novel gels include colloidal dispersion gels (CDGs), preformed particle gels (PPGs), SMG microgels, and temperature activated polymers (TAPs). These novel gel systems are either partially or wholly preformed agents with various size, chemistry, and properties. Since these novel gels are preformed, the issue related to cross-linking at reservoir condition is minimized. However, the reservoir conditions such as temperature, pH, salinity, permeability, adsorption, and heterogeneity still affecting the performance of novel polymer gel systems. In the following sections, the

properties, development, field application, and other relevant literature information related to these novel polymer gel systems are reviewed.

4.2.1 Colloidal Dispersion Gels (CDGs)

The Colloidal Dispersion Gel (CDG) system; developed by Tiorco Inc., consists of low concentration of a partially hydrolyzed polyacrylamide (100-1000 ppm) with moderate to high-molecular-weight (> 22 million Daltons), and a chelated aluminum citrate (or chromium citrate) solution as cross-linker (Castro et al., 2013; Ranganthan et al., 1998; Mack and Smith, 1994; Diaz et al., 2008). The typical polymer/cross-linker ratio reported in the literature is in the range of 20:1 to 100:1, and the typical concentrations used are 300 ppm polymer and 15 ppm Al^{3+} (Spildo et al., 2009; Spildo et al., 2010; Ranganthan et al.,1998). Because this system is composed of low concentration of polymers and cross-linkers, the bulk gel (continues network) cannot form and separated almost spherical micro-scale-gel particles (colloids) with the size range of 1-150 nm are created instead (Spildo et al., 2009; Castro et al., 2013).

The main characteristic of the colloidal dispersion gel system distinguishing it from bulk gel systems is that intermolecular cross-linking reactions are not dominant. Instead, the intramolecular cross-linking reactions dominated this system. Figure 4.8 shows the difference between bulk gel systems (intermolecular cross-linked) and CDG (intramolecular cross-linked) (Mack and Smith, 1994; Diaz et al., 2008; Diaz et al., 2015).

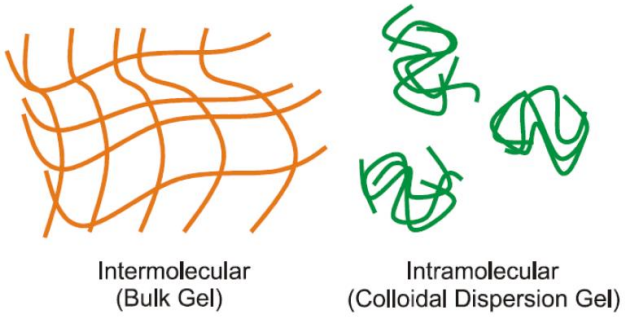


Figure 4-8: Difference between bulk gel and colloidal dispersion gel (CDG) (reprinted with permission from Daiz et al., 2008).

This system has been claimed to be slow forming and to be able to produce long-term, in-depth permeability modification in some mature water-flooded matrix (ordinary permeability) heterogeneous oil reservoirs with the maximum temperature of 94°C and total dissolved solids of 30,000 ppm (Mack and Smith, 1994). Coste et al. (2000) specified an upper temperature and salinity limits of 90°C and 5000 ppm, respectively. Spildo et al., (2010) reported the preparation and propagation of CDGs in total dissolved solid (TDS) of 35,000ppm and temperature of 85°C in a core flood test using real sandstone cores from a North Sea oil field.

It is speculated that gel aggregates are formed, which are then filtered from the brine solution by porous media, therefore providing resistance factor and residual resistance factor. These hypotheses are based on the interpretation of some field results in which colloidal dispersion gels have been successfully implemented (Mack and Smith, 1994; Fielding et al., 1994; Diaz et al., 2008; Diaz et al., 2015; Castro et al., 2013; Manrique et al., 2014; Leon et al., 2018).

Mack and Smith (1994), for the first time, reported the successful application of CDGs system in 22 of 29 field projects in the Rocky Mountain Region, USA. They reported the ultimate oil recoveries above 40% OOIP, in highly heterogeneous matrix reservoirs. The incremental oil recovery observed in 22 successful projects ranged from 1.3 to 18.2% of OOIP. Fielding et al. (1994) also reported a decrease in water-oil-ratio (WOR) and incremental oil recovery of 5% the OOIP in the North Rainbow Ranch Unit in Wyoming, USA. Chang et al. (2006) reported the successful application of this system in sandstone reservoirs with low salinity and low temperature in the Daqing oil field in China. The average water-cut before treatment with CDGs was 95.2%, and post-treatment data showed the maximum reduction of 19.8% in some production wells. The incremental oil recovery of 10.5% was achieved, which was above the planned value of 9.0%.

The most recent successful applications of this technology are reported in different oil fields in Argentina and Colombia (Diaz et al., 2008; Diaz et al., 2015; Castro et al., 2013; Manrique et al., 2014; Leon et al., 2018). Figure 4-9 clearly shows the improvement of injection profile created by the application of CDG system in the Loma Alta Sur oil field in Argentina. In an injection well of the Loma Alta Sur oil field, the layers showed more

uniform profile after treatment. Manrique et al. (2014) reviewed and summarized 31 implemented and ongoing CDGs projects in the US, Argentina, and Colombia since 2005. They concluded that the CDGs system could propagate in the reservoir without injectivity reduction problem. They further mentioned that the CDG system can be more economical in increasing oil recovery than regular polymer flooding; because substantially less amount of chemicals (polymers) are required.

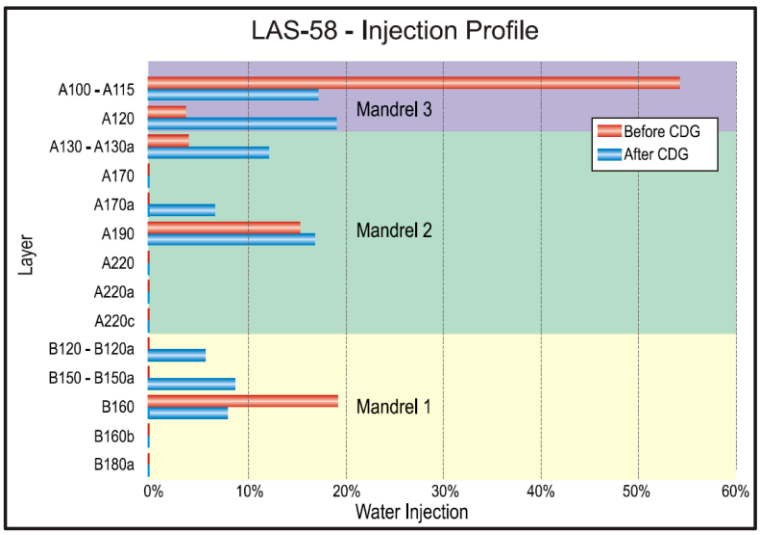


Figure 4-9: Injection profile improvement after CDG application in Loma Alta Sur Oil Field (reprinted with permission from Diaz et al., 2008).

Despite the successful results reported in some field applications, CDGs have uniquely gained longstanding controversial issues based on some laboratory evidence (Seright et al., 2006; Seright et al., 2015; Rao et al., 2017; Al-Assi et al., 2009). These controversies are mainly about the ability of CDGs to provide resistance factor and residual resistance factor deep into the reservoir and the effectiveness of this technology over regular polymer flooding.

Mack and Smith (1994) claimed that low concentration of Al^{3+} should prevent the chromatographic separation of the polymers and cross-linkers because cationic cross-linker tends to be more associated with anionic partially hydrolyzed polymer than the cationic rock surface. On the contrary, through laboratory investigations, Ranganathan et al. (1998) demonstrated that the retention and chromatographic separation of aluminum would reduce the colloidal dispersion gel treatment process to a regular polymer flood process. As shown

in figure 4-10, during the injection of colloidal dispersion gel solution prepared by 300 ppm polyacrylamide and 15 ppm aluminum citrate into 4ft long core with ordinary permeability of 3-4 darcy the concentration of aluminum collected in effluents was varying between 0-10 pm, while the initial polymer concentration (300 ppm) was observed in the effluent sample after around 2 PV injection.

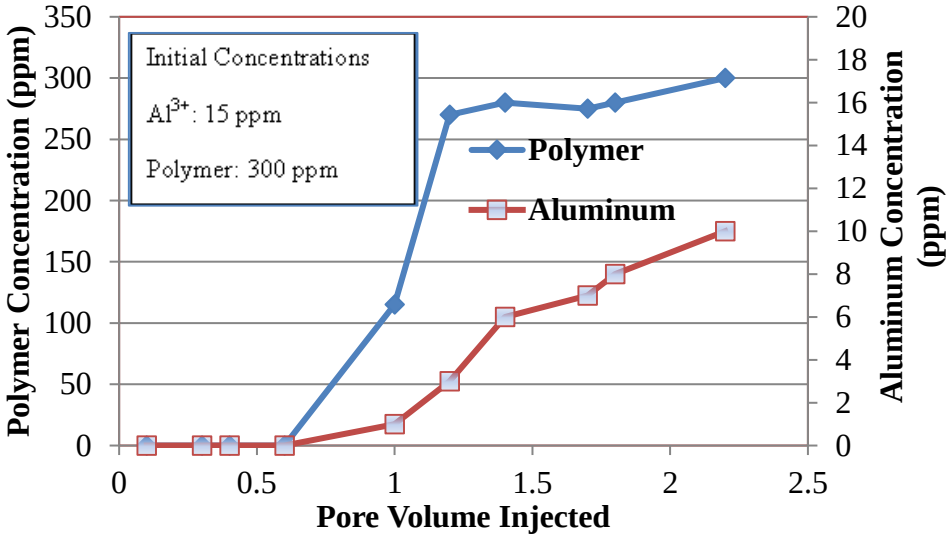


Figure 4-10: Polymers and aluminum concentration change (data points visually selected from Figure 7 in Ranganthan et al., 1998).

Mack and Smith (1994) also claimed the in-depth placement of colloidal dispersion gel system in porous media. They mentioned that due to the shear thinning behavior of CDGs solution, the application of this technology is not restricted by injectivity problems. They also introduced the term “transition pressure” to further describe the propagation of CDGs in porous media and to provide a tool for quantitative evaluations in the field design. In the lab conditions at transition pressure (differential pressure) range between 0.017 and 0.13 MPa the CDGs solution propagates through the pack of 100-mesh screens. They concluded that in field applications both the shear thinning behavior of the solution and high differential pressure near the wellbore provide the possibility of deeper placement. The injectivity and ability of in-depth propagation of CDGs system have been questioned in the literature. Al-Assi et al. (2009) are among those that question the in-depth propagation of CDGs in ordinary permeability reservoirs (reservoirs without fractures).

They showed that the propagation of this system, at an interstitial velocity of 5 ft/day, inside of a 10 D permeability porous media is limited to 12 feet.

Smith et al., (2000) through some lab studies concluded that gelation time of CDGs could be delayed by weeks or months if the gelant is injected at high velocity near the wellbore. They further mentioned that the injection of a freshly-made CDG gelant could develop high values of resistance factor far into the formation. Seright, (2006b) opposed these conclusions and set up some experiments (all at the temperature of 41°C) to further shed light on the propagation of CDG system in porous media.

First, they injected a solution of 300 ppm polymer (HPAM) without crosslinker into a 493 mD Berea sandstone core with the length of 0.43 feet at a high velocity of 143 ft/day. Internal pressure taps were installed to measure the resistance factor at a different section of the core. As the left hand of Figure 4-11 shows, after 118 PV of polymer injection, the resistance factors were almost equal for both core sections. They later injected the collected effluent from the above experiment into a 4 ft long Berea sandstone core with a permeability of 234 mD at a lower velocity of 2.7 ft/day. Four pressure taps were equally distanced to measure the resistance factors at five different core sections. As left hands of Figure 4-12 shows, again almost similar resistance factors (around 7) were observed in different core sections.

The second set of core flood experiments were implemented with a solution of freshly made (21 minutes old) CDGs solutions. The solution made of 300 ppm polymer and 15 ppm Al (Citrate) injected into the 493 mD Berea sandstone core with the length of 0.43 feet at a high velocity of 143 ft/day. As right hands of Figure 4-11 shows, the resistance factor at the first section of the core raised to more than 115 after 118 PV of injection while the resistance factor of around 21 was observed at the second section of the core. Similar to the regular polymer flood experiment, they injected the effluent collected from this core flood into the 4 ft long Berea sandstone core with permeability of 234 mD at a lower velocity of 2.7 ft/day. The CDGs system at the time of injection was 2.7 hours old (~136 minutes). As middle of Figure 4-12 shows, after some time the resistance factor in the first section of the core raised to very high value while the value of resistance factors in the other four sections were very low.

Seright, (2006b) repeated the above set of experiments with different values of core permeability, injection rate, and core lengths and concluded that when the CDGs aggregates grow to the size of the pore throat, they stop propagating into the formation. They also mentioned that gelant propagation for the time of weeks or months, as suggested by Smith et al. (2000), are not achievable with this technology.

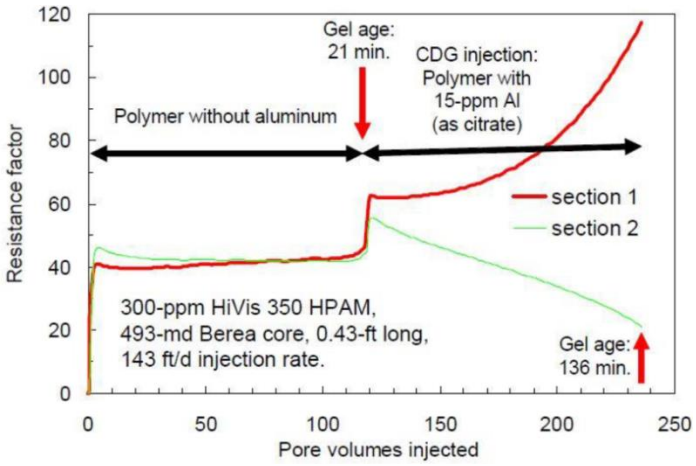


Figure 4-11: Resistance factor in 0.43 ft long Berea core during high rate injection (Seright, 2006b).

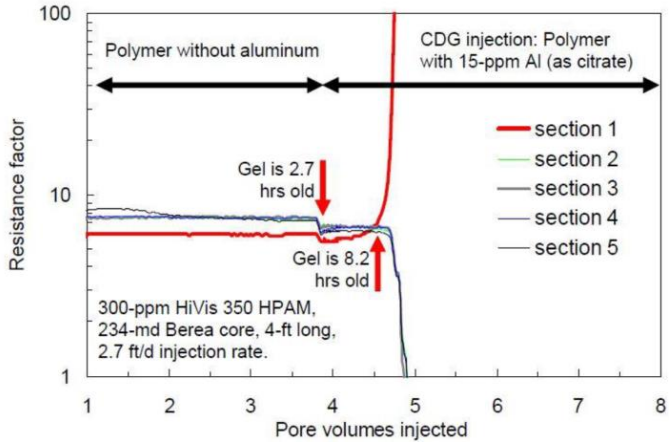


Figure 4-12: Resistance factor in 4 ft long Berea core during low rate injection (Seright, 2006b).

Manrique et al. (2014) and Diaz et al. (2015) most recently examined the CDGs flood in pilot and field applications. They both used Hall plots to investigate the effectiveness of this technology in improving conformance. They each separately claimed that CDGs could provide higher resistance and/or residual resistance factors compare to

regular polymer flooding. Seright et al. (2015) and Rao et al. (2017) each offer a complete document to question the mentioned claims by CDGs vendor. Seright et al. (2015) have examined 24 published papers including: 16 papers advocating the effectiveness of CDGs and 8 articles questioning the CDGs technology. In part of their conclusion, they mentioned that Hall plots should not be trusted as tools to quantify the resistance and residual resistance factors, because Hall plots only monitor the injection pressures at the wellbore and cannot differentiate formation damage, face plugging, fracture extension from mobility and conformance improvement.

Similar work has been recently published in 2017 (for partial fulfillment of the requirements for the Master of Science degree) and questioned the vendor claims (including Diaz et al., 2015) about the superiority of CDGs over polymer floods (Rao et al., 2017). They concluded that the money spent on the cross-linking of the CDGs is a waste and there is no evidence that CDGs can provide better resistance factor or/and residual resistance factor than regular polymer floods. They also mentioned, a similar conclusion made by Seright et al. (2015), that the CDGs may damage the formation production by face plugs, excessive fracture extension and excessive loss of polymers.

4.2.2 Preformed Particle Gels (PPGs)

Preformed Particle Gels (PPGs) are millimeter-sized, salt-tolerant, temperature-resistance pre-crosslinked, and highly swellable gel particles developed by PetroChina in 1996 (Coste et al., 2000). The initiation of this technology was motivated by the shortcomings of traditional bulk and colloidal dispersion gels to provide in-depth flow diversion under the condition of high temperature, high salinity and severe channeling in the reservoirs (Coste et al., 2000; Bai et al., 2007; Bai et al., 2004).

The Preformed Particle Gels (PPGs) are dried, crushed, and sieved particles that can absorb brine and swell up to 200 times in size (Figure 4-13) (Bai et al., 2007; Bai et al., 2004). The particles can form a stable suspension in brine and inject into the formation. The swelled particles, having elastic/deformable nature, can travel deep into the formation where they can fully or partially block the high permeability channels/fractures and thus providing conformance control (Coste et al., 2000; Bai et al., 2007; Bai et al., 2004). The Swelling Capacity (A) is defined as follows:

$$A = \frac{Ml - Ms}{Ms} \quad (4.2)$$

where M_l and M_s are volumes of the particle gel after and before swelling, respectively.

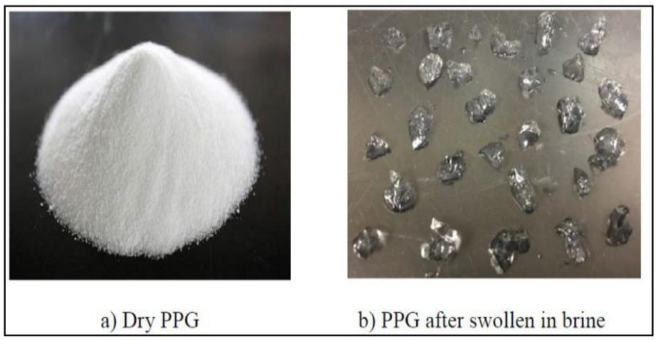


Figure 4-13: PPG before and after becoming fully swollen (reprinted with permission from Imqam et al., 2017).

The preparation of PPGs has different steps. First, a bulk gel forms by a solution polymerization method of an acrylamide monomer (AM), a cross-linker (e.g., N, N'-methylenebisacrylamide), an initiative (e.g., sodium peroxydisulfate) and other additives (e.g., bentonite clay) at room temperature. Then, the bulk gel mechanically cuts into small pieces and dries at a higher temperature to form solid particles. Finally, the dried particles sieve/screen to obtain the required size for specific field applications (Bai et al., 2007; Bai et al., 2004). Figure 4-14 is a schematic of the PPGs preparation procedure.

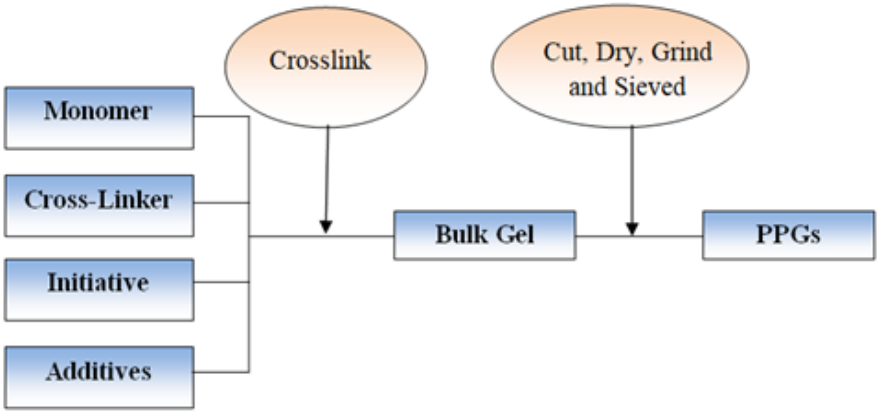


Figure 4-14: Preformed particle gels fabrication procedures (adopted from Bai et al., 2004).

Preformed particle Gels (PPGs) are useful for most types of brines (salt concentrations) and can resist high reservoir temperature (Coste et al., 2000). The results of lab experiments show that PPGs can withstand a temperature of 120°C for one year (Bai et al., 2007). The high salinity resistance of up to 300,000 ppm TDS reported in the literature (Bai et al., 2004). Field application of this technology also confirmed the high salinity and temperature stability of this gel system. For example, treatments of two injector wells at Zhongyuan field in China proved the effectiveness of this technology at high salinity (150,000 ppm) and high temperature (107°C) reservoir where after 3 months of the treatment 3,239 tons of additional oil was produced (Bai et al., 2004). Another advantage of preformed particle gels (PPGs) is the absence of chromatographic separation, which makes them suitable for in-depth flow diversion (Bai et al., 2007; Bai et al., 2004).

The propagation of swelled PPGs in the porous media is controlled with some parameters such as the size of the particles, deformability of the gel particles (gel strength), the permeability of the reservoir (pore throat diameter) and the injection flow rates (Bai et al.,2007; Bai et al., 2004). Based on the experimental studies on the transport of PPGs under different conditions, various flow patterns of PPG in porous media are possible (Figure 4-15), which are listed below:

- Direct Pass: When the PPG is so small compare to the pore throat, it can easily displace with the injected water.
- Adsorption: Small PPG can also be adsorbed onto the rock surface.
- Trap: When the particle is both large in size (compare to pore throat diameter) and dense (have high strength), it can mechanically block the pore throat.
- Deform and Pass: When particle is larger than the pore throat but has elastic nature, it can pass the pore throat, due to the force applied with injected water, and recover to its original size.
- Shrink and Pass: When the particle is larger than pore throat, it can shrink in size by dehydration and pass the pore throat.
- Snap-off and Pass: When particle is larger than the pore throat and broke into smaller particles while pass the pore throat (Bai et al., 2007; Bai et al., 2004).

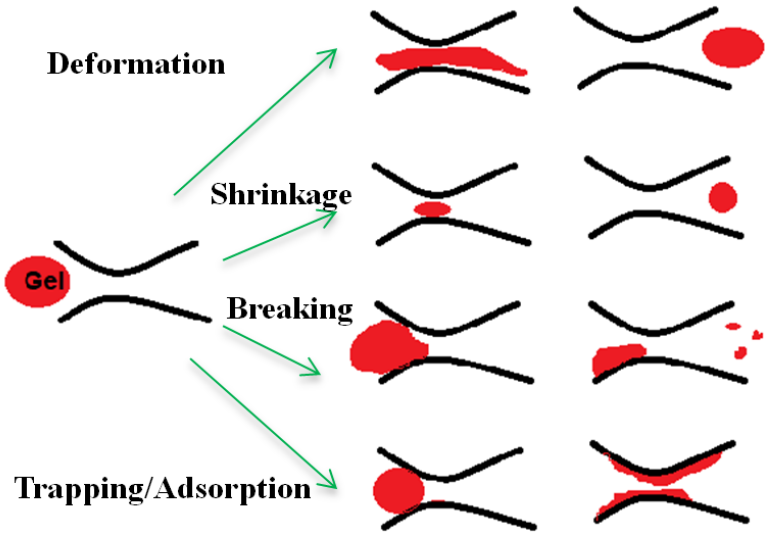


Figure 4-15: Mechanisms of PPGs passing through pore throats (adopted from Bai et al., 2007).

Therefore, the propagation of millimeter-sized PPGs into the porous media can be restricted by rock permeability. Sang et al. (2014) mentioned that the millimeter-sized particles could not be injected into the formation unless it has a permeability of several Darcy. Elsharafi and Bai, (2015) reported the minimum permeability of 0.3 Darcy. They also mentioned that for the swollen PPG to penetrate into the formation, the ratio of PPG size to pore throat should not exceed 17. The exact permeability for the application of PPGs is not clear at this point, but field application and laboratory core-floods suggested that the PPGs are applicable for fracture reservoirs and also formations that have been flooded for several years (Bai et al., 2007). The successful field applications of PPG have been reported in both mature sandstone and fractured reservoir in China (Qiu et al., 2014). Qiu et al. (2014) reported the successful results of 655 treatments with PPGs without any injectivity problems. The absence of the injectivity problems of these particles may be attributed to the elastic nature of the swollen particles and also the shear thinning behavior under the high flow rates. The core flood experiments also confirm the reduction of Fr by flow rate (Bai et al., 2007; Saghafi et al., 2016a).

Another advantage of PPGs is the possibility of selective penetration into the high permeability zone and minimizing the damage to low permeability oil bearing zone by controlling the properties of the particles. The selectivity of PPG is only suitable for high

permeability contrast between layers (Imqam et al., 2014). Elsharafi and Bai, (2015) demonstrated that both strong (DQ gel) and weak (LiquiblockTM 40K) PPGs with small size (100-120 mesh) damage the low permeability cores with 5-25 mD while the large, strong PPGs (30-80 mesh) do not damage the low permeability cores. They also mentioned that, when the particle gels swelled in low concentration brine, more damage can occur.

The Relative Permeability Modification (RPM) of PPGs also been proved through lab experiments. The shrinkage and dehydration of swollen gels under the oil capillary will reduce the gel size and consequently decreases the residual resistance factor. Imqam et al., (2014) demonstrated that the fracture filled with PPGs could experience F_{rrw} of 100-1700 higher than F_{rro}. They also mentioned that the gel strength is the most critical factor for controlling the RPM property of the gel and softer gel swelled in lower brine concentration can provide better RPM effect.

The effects of brine concentration and pH in the performance of PPGs have been studied recently. Bai et al. (2007) shows that basic pH does not influence the swelling capacity of PPG while acidic pH, especially below 6, reduced the swelling capacity by 2 - 3 factors. Imqam and Bai, (2015) showed that the lower brine concentration results in more particle swelling and less strength, while higher brine concentration provides smaller swelling gels with higher strength. They also conclude that for better plugging of the high permeability zones, larger pre-activated particle gels and higher brine concentration is better than the small pre-activated particles with lower brine brine concentration. Saghafi et al., (2016a) also supported these conclusions. They demonstrated that increasing the size of pre-swelled PPGs from 37-44 to 74-105 µm increases the F_{rrw} from 29 to 79. From the above discussions, for better permeability reduction of the high permeability zones, the larger pre-swelled PPGs with higher brine concentration is more effective while for relative permeability reduction (RPM) elastic PPGs swelled in low brine concentration should be considered.

Preformed Particles Gels (PPGs) have been recently gaining more attention and several researches conducted to improve the plugging efficiency and stability of this system. To further enhance the plugging efficiency of PPGs in fractures, Zhang et al. (2019) used PPGs and HPAM/Cr^{3+} to plug a fracture efficiently. They demonstrated that

the combination of both systems could significantly increase the oil recovery (29%) compare to using PPGs alone (18.5%). Alhuraishawy et al. (2017) mentioned that the PPG system composed of mixing size particles could more effectively plug the fracture that uniformed sized PPGs. Both of the methods were proposed to reduce the internal permeability of the PPG gel pack inside the fracture. In the same context, Pu et al. (2018b) proposed a novel Re-Assembling Preformed Particle Gels (RPPG) system (Figure 4-16). In this system, the swelled, individual preformed particle gels can re-assembly to form a bulk gel. The authors also tested the long-term thermal stability, at the experiment temperature of 45°C, of the novel system and showed that RPPG can be stable for 300 days.

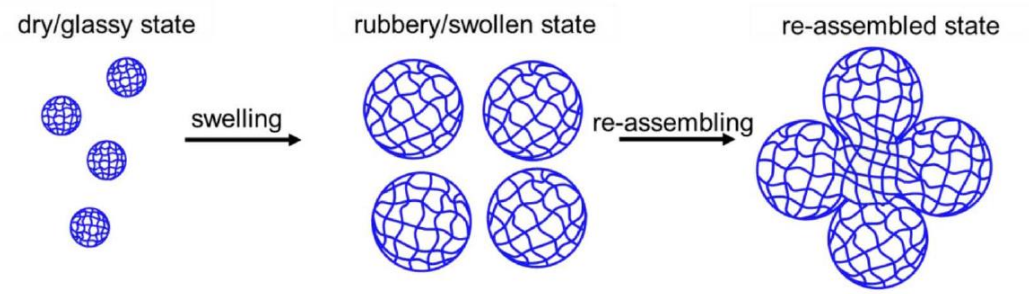


Figure 4-16: The RPPG re-assembling procedures (reprinted with permission from Pu et al., 2018b).

To further improve the thermal stability and strength of PPGs, several authors conducted the application of nanotechnology. In this context, laponite XLG nanocaly (Tongwa and Bai, 2015), nano fly ash (Kumar et al., 2019), starch and sodium montmorillonite (Long et al., 2019; Saghafi et al., 2016b) were added to PPGs system recipe to improve its properties further. Saghafi et al. (2016b) reported that the PPG synthesized by sodium montmorillonite can resist temperature of up to 145 °C and Long et al. (2019) demonstrated that starch -PPG had thermal stability of up to 187°C, but the long term stability of these new PPG recipes is not disclosed in the literature.

4.2.3 Microgels

Microgels are micrometer-size pre-cross-linked polymeric aggregates, which are strength-adjustable, size-controllable, stable, and nontoxic and behave like large polymer molecules (Zaitoun et al., 2017). Chauveteau et al. (1999) proposed the first microgels

under the name of "STARPOL" for water-shut off applications. The STARPOL microgels were synthesized by cross-linking of an acrylamide-based polymer solution with zirconium lactate under shear stress. Chauveteau et al. (2004) later reported a new type of nontoxic microgels based on the cross-linking of acrylamide monomer (AM) and sulfonate (AMPS) monomers with an organic, nontoxic, and neutral cross-linker. As shown in Figure 4-17, the new types of microgels referred to as SMG microgels with two different sizes of 0.3 and 2 µm and various chemistries and cross-linking densities are recently reported in the literature (Zaitoun et al., 2007; Dupuis et al., 2013; Zaitoun et al., 2017). This type of microgels is synthesized by water-in-oil emulsion polymerization (Zaitoun et al., 2007). During the manufacturing process of SMG microgels, different parameters such as chemistry, consistency, and size can be varied to be suitable for various application and conditions (Zaitoun et al., 2017). SMG microgels can be produced with high cross-linking density to form hard microgels for water shut off applications or with low cross-linking density to form soft microgels for in-depth flow diversion applications (Zaitoun et al., 2007). The application of the new commercially available SMG microgels systems is not restricted to water-shut off applications, and recently they have been used for conformance control in injector wells as well (Zaitoun et al. 2017).

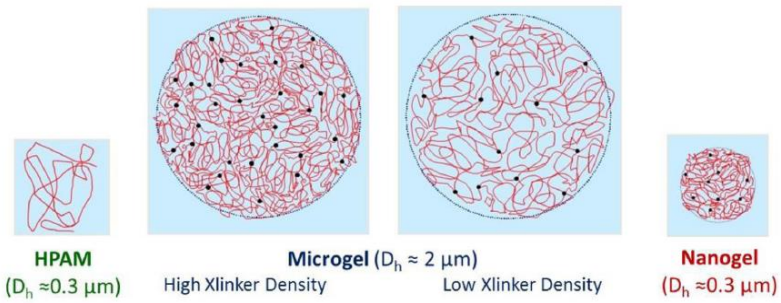


Figure 4-17: Different SMG microgels (reprinted with permission from Zaitoun et al. 2017).

The mechanism of permeability reduction by microgels is explained by the monolayer or multi-layer adsorption onto the porous rock surface (Chauveteau et al., 2004). The microgels can be synthesized to be either attractive or repulsive to each other (Chauveteau et al., 2004). The interaction between microgels can be expressed through the

term Huggins Constant (K_H). When the K_H<0.3 the microgels are repulsive, forming monolayers onto the rock surface and are suitable for in-depth flow diversion and when K_H>0.3 microgels are attractive, creating multi-layers onto the rock surface and are ideal for near wellbore water-shut off application. Therefore, with changing the chemistry and varying the Huggins constant different penetration depth can be achieved with microgels systems (Chauveteau et al., 2004). For soft repulsive microgels, the permeability reduction (F_{rr}) is due to the adsorption of the monolayer of microgel onto the rock surface (Cozic et al., 2009).

Microgels are known to have excellent mechanical, thermal, and chemical (Salinity) stabilities (Chauveteau et al., 2004; Cozic et al., 2009; Dupuis et al., 2013). Dupuis et al. (2013) studied the thermal and mechanical stability of different microgels with varying cross-linking densities. They demonstrated that the microgels are more stable than traditional polymers when exposed to mechanical shear stress. In their experiment, microgels could resist mechanical shear rates up to 1.2× 106 s^{-1}. The thermal stability of microgels was also evaluated at 85, 105, and 140°C. The microgels with high cross-linking density showed minimal thermal degradation after one-month aging at 140°C (Dupuis et al., 2013). Cozic et al. (2009) studied the effect of salinity on the stability of adsorbed microgels mono-layers and showed that even salinity equal to 200,000 ppm TDS has no impact on the hydrodynamic thickness of mono-layers of microgels adsorbed on the rock surface.

Microgels are also known to have relative permeability modification and selective water permeability reduction properties (Rousseau et al., 2005; Dupuis et al., 2015). Dupuis et al. (2015) mentioned that microgels are relative permeability modifiers (RPM). They treated a core with a permeability of 3.8 Darcy with 2µm microgels. The oil permeability does not change at all, while water permeability significantly reduced (Dupuis et al., 2015). Soft Microgels can shrink under the influence of oil-water capillary pressure and prevent oil permeability reduction while the adsorbed mono-layer of microgels can reduce the permeability to water significantly with reducing the effective size of pore throats (Rousseau et al., 2005). Rousseau et al. (2005) also compared the permeability reduction of a solution with 3000 ppm of microgels injected in a Berea core with a permeability of

0.3 D at both residual oil and water saturation. They concluded that adsorption of the microgels onto the pore surface severely reduced the water permeability while oil permeability at residual water saturation remains unchanged. Figure 4-18 shows the difference between permeability reduction of microgels to water and oil at residual oil saturation (S_{or}) and residual water saturation (S_{wi}) (Zaitoun et al., 2007).

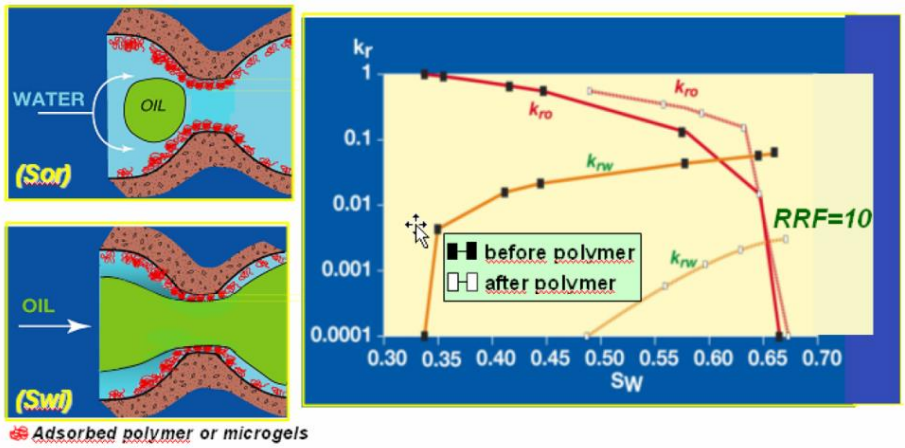


Figure 4-18: Relative permeability modifications by microgels adsorption (reprinted with permission from Zaitoun et al., 2007).

The low viscosity of microgel solutions and the possibility of manufacturing in narrow size distribution make these agents to be suitable for selective permeability reduction of high permeability zones without penetration in low permeability zones (Yao et al., 2012; Yao et al., 2016). Zaitoun et al. (2007) demonstrated that when a solution of microgels with a concentration of 3,000 ppm injected in a core with a permeability of 205 mD, different Fr (Resistance Factor) were obtained for different flow rates. They showed that Fr values of 30 and 50 could be obtained at flow rates of 200 $cm^3\ h^{-1}$ and 20 $cm^3\ h^{-1}$, respectively. This observation showed that microgels are having shear thinning behavior. Therefore, they can penetrate deep into the porous media when injected under higher flow rate. Yao et al. (2012) mentioned that for microgels to be able to penetrate to the sand pack, the ratio of particle/pore throat size should be smaller than 3.25. Yao et al. (2016) experiments showed that microgels reduced the permeability of 3.642 D core to 0.546 D while the permeability of 0.534 D core reduced to only 0.512 D. Chauveteau et al. (2004) also simulated the selective permeability reduction of microgels with average size of 1.5 µm in a well with three layers having different permeability (K=1000, 100 and 75 mD). As

Figure 4-19 shows, microgel solution can selectively penetrate into the highest permeability layer.

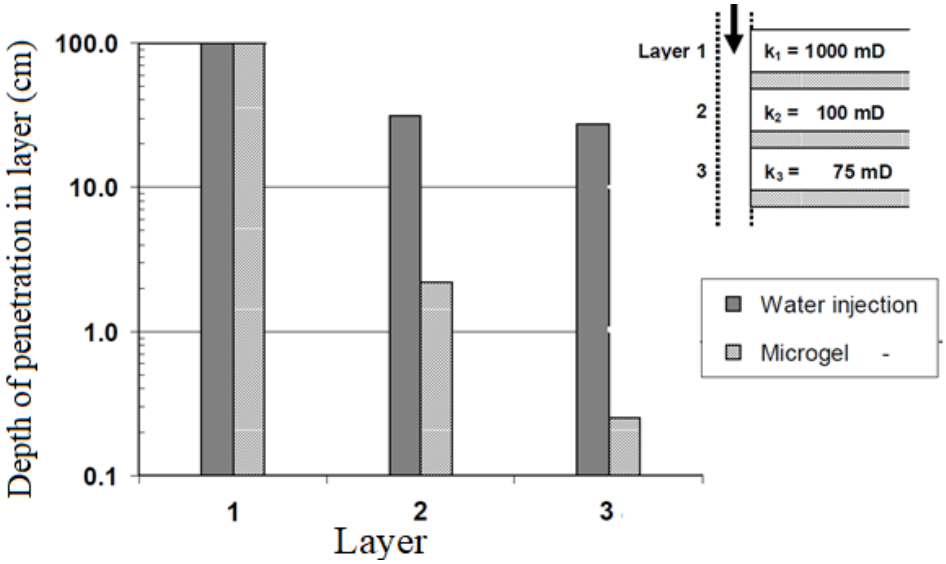


Figure 4-19: Depth of penetration in layers with different permeability when microgels with a diameter 1.5 µm reached 100 cm in high permeability layer (reprinted with permission from Chauveteau et al., 2004).

Successful field applications of microgels were also reported in the literature. Zaitoun et al. (2007) reported the first water-shut off application of SMG microgels in underground gas storage well. The well was producing with 100% water cut due to the existence of a thief zone with a permeability of 6 Darcy with adjacent layers having low average permeability of 205 mD. The well was treated with the bullhead injection of 26 m^3 of a solution containing 6000 ppm of 2 µm soft microgels. As a result of the treatment, water production reduction in the range of 3 to 5 times, and significant improvement in gas production was observed. A similar positive result was recently found in an Omani oil reservoir. The producer well was at 100% water cut. Since the precise identification of the water-bearing zone was not possible, 2 µm soft microgels solution having RPM effect was selected for bullhead injection of the whole strata. As a result of the treatment, the water cut was reduced to 85%, which consequently produced 9,000 bbls of incremental oil in one year (Dupuis et al., 2015).

Dupuis et al. (2016) recently reported the application of SMG microgels in an injector well surrounded with seven producers in and Omani Oil field. The reservoir is heterogeneous sandstone with the permeability ranged between 10 and 1000 mD with an average permeability of 125 mD. The reservoir temperature and salinity were measured to be 48°C and 8,000 ppm, respectively. During three months, 10,000 m^3 of SMG microgels solution with the concentration of 500 ppm and a size of 2 µm were injected into the injector well. As a result, after one-year water production was reduced by 125,000 bbl while oil production has increased by 10,000 bbls. Another conformance control with SMG microgel was recently reported in a heterogeneous sandstone reservoir in the same Omani oil field. The reservoir with an average permeability of 200 mD (ranged between 10-1200 mD) was treated with the bullhead injection of 9,000 m^3 of 2 µm SMG microgels. After 26 months, more than 5440 m^3 incremental oil has been extracted, thus the economics of the project evaluated as 0.8 lbs/bbl (0.8 Ibs of raw material for 1 bbl incremental oil production) (Zaitoun et al., 2017).

For the field application mentioned above, the microgels system has to be delivered on the field in the form of inverse emulsion; therefore, additional transportation and handling cost were required (Bai et al., 2015). Microgels size and chemistry need to be selected based on petro-physical properties of the target formation such as porosity, permeability and rock type; therefore, application of this technology required specific screening criteria and cost considerations.

4.2.4 Thermally Activated Polymers (Bright Water)

Thermally activated polymers/particles (TAPs) are sub-micron-gels developed by a collaborative industry research project among BP (British Petroleum), Chevron, Texaco and Nalco in 1997 (Frampton et al., 2004; Pritchett et al., 2003). This technology further commercialized by Tiorco-Nalco under the name of "Bright Water." The main objective of this collaborative effort was to introduce a highly swellable and time-delayed material to plug the high permeability zones deep in the reservoir and to improve the injection profile by diverting the injected chase water-flood into less permeable un-swept oil bearing zone (Frampton et al., 2004; Pritchett et al., 2003).

Thermally activated particles (TAPs) are sub-micro-sized-particles (0.1-1 µm) consist of sulfonated polyacrylamide (AMPS) polymer chains cross-linked with both labile (reversible) and non-labile (stable) cross-linkers prepared in mineral oil (Trasher et al.,2016; Fabbri et al., 2015). This system is synthesized by inverse emulsion polymerization process which is the preferred method to achieve a suspension of narrow size range sub-micro-particles. The suspension of TAPs later will mix with a surfactant prior to injection to disperse the particles into the injection water and consequently prevents the conglomeration of them during propagation in the reservoir (Bai et al., 2015).

In porous media, un-swelled thermally active sub-micron-gels often referred to as "Kernels," are injected with water having a low temperature compared to the reservoir (Pritchett et al., 2003). The temperature difference between injected water and reservoir creates a so-called "Thermal Front" somewhere deep in the reservoir (Fabbri et al., 2015). The cold water containing kernels have a low viscosity (close to water viscosity), and as a result, selectivity moves to high permeability layers (thief zones) (Pritchett et al. 2003; Garmeh et al., 2011; Roussenance and Toschi, 2010). As Figure 4.20 shows, the sub-micron-particles slowly adsorb heat from the reservoir, and at a specific temperature and time (after reaching the thermal front), the kernels pop like "popcorn" and increase in volume (Frampton et al., 2004; Garmeh et al., 2011). The key feature of these kernels is their thermo-responsive property. When reached the thermal front and exposed to elevated temperature in the reservoir, the labile cross-linkers in the system go through dissociation. Therefore, the particles, now having less dense cross-linking structure, start to absorb the surrounding brine and swell. The non-labile cross-linkers, on the other hand, maintain the integrity of the expanded particles and also control the final size of the particles.

These kernels can swell by 10 times when brine adsorbed to their structure (Garmeh et al., 2011). Therefore, it leads to the plugging the pore throats in thief zones and increasing in the apparent viscosity of the solution; thus increasing the F_{rr} (Frampton et al., 2004; Thrasher et al., 2016). As Figure 4-21 shows, the injected water consequently moves to un-sweep oil-bearing zone, where it can recover additional oil (Roussenance and Toschi, 2010; Husband et al., 2010).

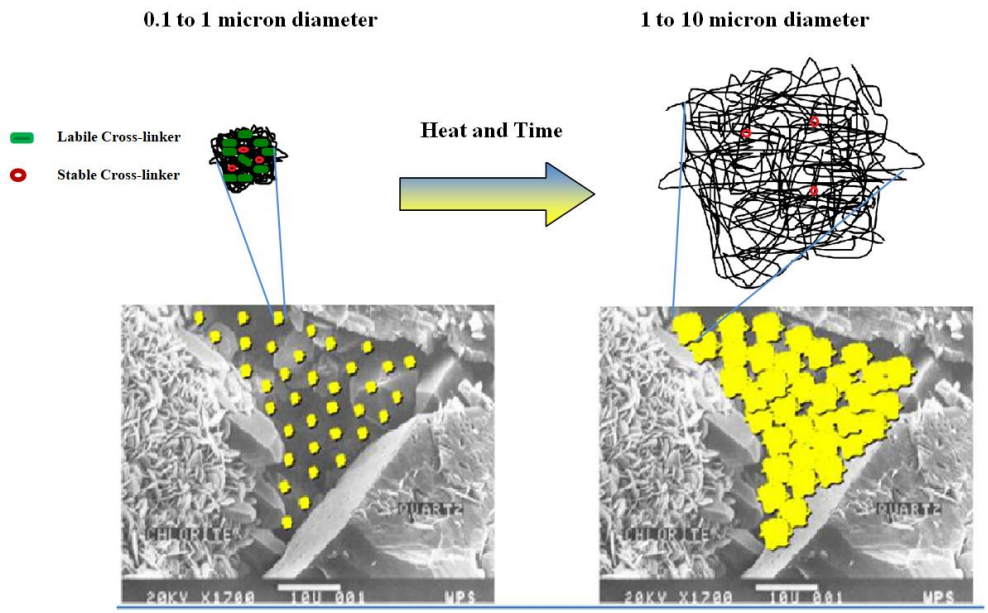


Figure 4-20: Bright Water activated by heat and time (reprinted with permission from Garmeh et al., 2011).

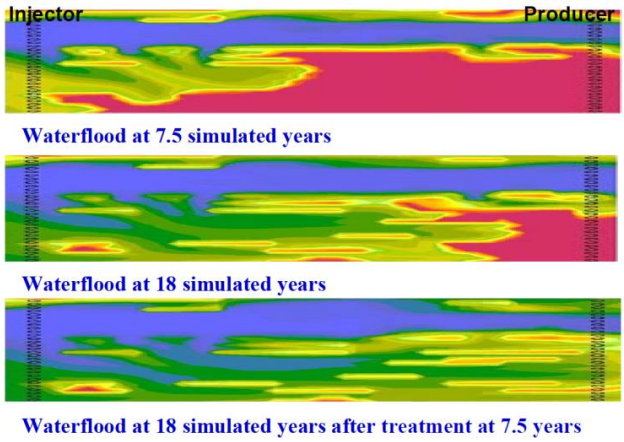


Figure 4-21: Simulation of vertical sweep improvement by Bright Water (reprinted with permission from Husband et al., 2010).

The injectivity, propagation and plugging efficiency of Bright Water have been studied by several authors (Pritchett et al., 2003; Frampton et al., 2004; Mustoni et al., 2010; Garmeh et al., 2011). Pritchett et al. (2003) stated that the kernels do not have injectivity problems because they are only one component at the time of injection and therefore chromatographic separation could not occur. Frampton et al. (2004) demonstrated that the sub-micron-particles could penetrate through sandpack without face plug. They

also mentioned that slug of the Bright Water exhibits low F_r. Therefore, it can selectivity propagate deep into high permeability pathways. Mustoni et al. (2010) sandpack experiments show a similar result. As Figure 4-22 shows, the 40 ft long sandpack treated with 3,000 ppm TAPs solution followed by post-water injection does not experience flow resistance in the first 10 ft of the core. While the other three sections of the core experience high F_{rr} due to the swelling of the TAPs after 200 days of aging time.

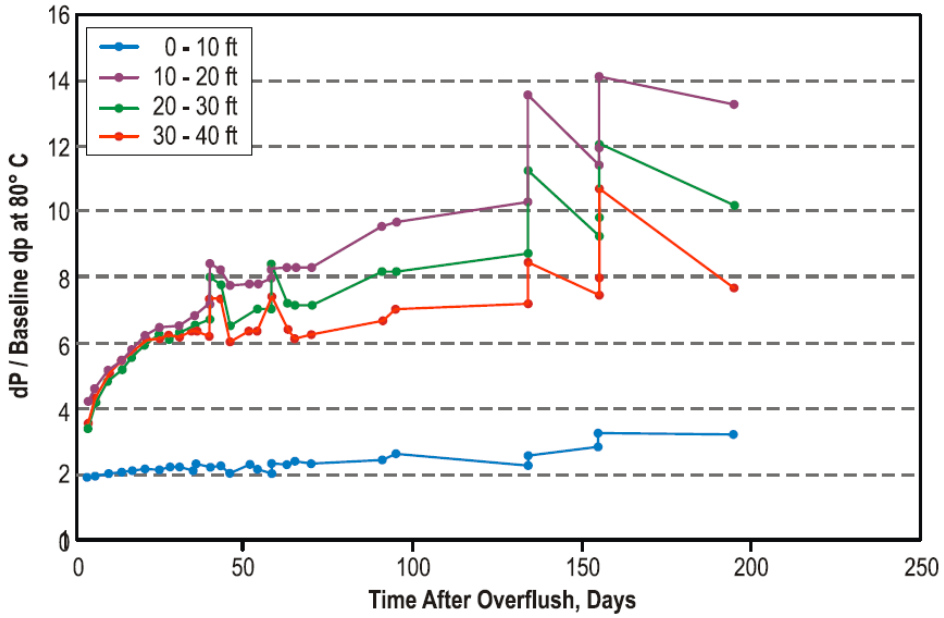


Figure 4-22: Sand pack test results: 3000 ppm Bright Water at 80° C (reprinted with permission from Mustoni et al., 2010).

Different authors also studied the ability of TAPs to create Frr under different circumstances (Frampton et al., 2004; Ohms et al., 2010; Garmeh et al., 2011; Fabbri et al., 2015). Frampton et al. (2004) mentioned that the ability of TAPs to create high F_{rr} is limited compared to bulk gels, but even low F_{rr} values are sufficient for in-depth flow diversion applications. Sandpack lab tests and simulation results indicate that injection of TAPs solution with the concentration of 1500 to 3500 ppm into the sand with a permeability of 560 to 670 mD can provide Frr values of 11 to 350 (Ohms et al., 2010). Garmeh et al. (2011) through lab test demonstrated the possibility of Frr reduction by washed-out. In their experiment, a sandpack with the permeability of 290 mD was treated with a solution of TAPs with a concentration of 5,000 ppm and aged for 50 days at the temperature of 30 °C. The maximum F_{rr} of 80 was observed during the chase flood, but it

reduced to less than 60 after only 3 PV of water injection. Most recently Fabbri et al. (2015) studied the performance of TAPs in a 7.5 D core. In their experiment, a core was saturated with TAPs solution and aged for 41 days at the temperature of 50°C. The F_{rr} value of 1.3 was observed which reduced to 1.1 after 13 PV of water injection.

The first trial field application of this technology as reported by Pritchett et al., (2003) also confirmed that low viscosity slug of Bright Water could be injected into the reservoir without any injection and propagation problems. This trial field application was conducted in Minas filed in Indonesia in 2001. During the trial, forty-two thousand barrels of a solution having 4500 ppm of micro-particles mixed with 1500 ppm of surfactant were pumped into the formation. The injection tracer test and pressure fall-off test confirmed that the sub-micron-particles could improve the injection profile without compromising the injection well by face plugging. The results also showed that sub-micron-particles could propagate considerable distance from the injector (125 ft).

First commercial application of Bright Water was successfully implemented in Milne Point oil field in Alaska with incremental oil recovery of 60,000 bbls of oil (Ohms et al., 2010). Husband et al. (2010) also reported the application of Bright Water in Prudhoe Bay oil field in Alaska. In this project, three injectors of the field were treated with the injection of 630-645 bbls of Bright Water accompanied by 310-335 bbls of surfactant. The results of the treatment showed the incremental oil recovery of 500,000 bbls of oil and 4% reduction in water cut. These promising results further encourage the operators to expand the size of the project in this oil field. Trasher et al. (2016) reported that up to 2014 more than 90 treatments were implemented in Prudhoe Bay oil field alone. The application of Bright Water was not limited to Alaska. Some successful field application of this technology is also reported in Argentina (Yanez et al., 2007; Mustoni et al., 2010), Brazil (Roussennace and Toschi, 2010), Gulf of Suez (Towns et al., 2013), Tunisia (Fethi et al., 2010) and most recently in Equatorial of Guinea (Choudhary et al., 2014).

The application of Bright Water in fields also helps the operators to understand the conditions under which this technology should be considered. Several authors through lab, field and simulation works develop different criteria for TAPs application (Pritchett et al., 2003; Yanez et al., 2007; Manrique et al., 2012; Thrasher et al., 2016).

- Early water breakthrough (at high water cut)
- Water channeling problem (with cross-flow with moderate to low Kv/Kh)
- Available oil in un-swept area
- Transit time greater than 30 days and placement half-way between injector and producer (Pritchett et al., 2003, Manrique et al., 2012). Izgec et al. (2012) mentioned that slug closer to producer is more favorable.
- Only sandstone reservoir and not suitable for carbonate reservoirs with fractures.
- High pH (greater than six) for both injection water and reservoir.
- Permeability contrast of at least 5 reported by Pritchett et al. (2003) while Thrasher et al. (2016) and Manrique et al. (2012) reported the permeability contrast of greater than 2 and 3, respectively.
- Temperature: Pritchett et al. (2003) reported temperature range of 50-150 ^{o}C while Manrique et al. (2012) specified 20-120 ^{o}C as the temperature range.
- Water salinity of 70,000 ppm and 150,000 ppm were reported by Pritchett et al., (2003) and Manrique et al. (2012), respectively while (Fethi et al., 2010) reported successful application of this technology up to the salinity of 260,000 ppm TDS.
- Thief zone permeability of at least 100 mD (Pritchett et al., 2003) and maximum 3.4 and 2.5 Darcy reported by Frampton et al., (2004) and Thrasher et al., (2016), respectively. Yanez et al. (2007) reported easy propagation of TAPs into matrix sandstone with permeability of 50 mD. Town et al. (2013) specified the range of 50 to 4000 mD. Choudhary et al., (2014) reported the successful application of this technology in thief zone with permeability of 25 Darcy.

Bai et al. (2015) mentioned that, because Bright Water is the combination of both surfactant and particles, it is difficult to distinguish the initial reason of the oil recovery improvement. However, no further article in the literature that argues the above statement was found. From the above discussions, that Bright Water technology showed satisfactory results in a good number of field applications especially when the conformance problem is

due to relatively low permeability thief zones in matrix reservoirs, but the exact range of permeability suitable for its application and other parameters still need to be studied.

4.3 Comparison of polymer gel systems

The six polymer gel systems discussed in this paper are compared based on their properties, their advantages, disadvantages, and types and location of field applications and summarized in Table 4-3. The important properties such as deep permeability reduction, relative permeability modification, gelation time and strength, selectivity of penetration into high permeability layers are among factors considered for comparison in Table 4-3. Furthermore, Table 4-4 can be referred for comparison of these technologies based on reservoir condition, such as reservoir temperature, formation water salinity, pH and thief zone permeability. The comparison of polymer gel systems provided in Table 4-3 and Table 4-4 could be beneficial to the operators and reservoir engineers in the selection of proper polymer gel systems. Conventional in-situ bulk gel systems: HPAM/Cr(III) acetate and PAtBA/PEI are mainly applied for near wellbore conformance control applications in both injector and producer wells. Novel microgels: CDGs, PPGs, SMG microgels and TAPs, mainly applied for in-depth flow diversion conformance control applications.

Table 4-1: Comparison of polymer gel systems: advantages, disadvantages and field applications (reference to information in table within the text).

Gel Category	Gel System	Descriptions	Advantages	Disadvantages	Field application
Conventional In-situ Bulk Gels	HPAM/Cr (Acetate)	• In situ bulk gel or partially preformed gel • High concentration polymer • Ionic intermolecular reaction • Widely applied system.	• RPM properties: F_{rrw} 100-1000 times higher than F_{rro} • Relatively low cost • Availability of chemical	• Low temperature and salinity resistance • Precipitation at basic pH • Chromatographic separation, diffusion, dilution affect gelation	• Mainly Water shut off & profile modification, In depth diversion in some fractures

	PAtBA/PEI	• In situ bulk gel • Covalent bonding • Nontoxic crosslinker • Controllable hydrolysis degree • Retarder and Strength Enhancer can be used	• High temperature resistance • No precipitation at high pH • strong sealant • Propagation better than HPAM/Cr	• Cannot be bullheaded • No RPM property • Not effective at acidic formation water • Cannot be used for in depth flow diversion	• Field applications Middle East, Mexico, etc. • Near wellbore water shut off, casing leaks, etc.
Novel Microgels	CDG	• Dispersion of gel aggregates formed in-situ • 1-150 nm sized colloids • Intramolecular reaction • Low concentration polymer 10-1000 ppm	• Low cost due to less chemical • High injectivity • Possibility of in-depth diversion • Large volume	• Low thermal and salinity resistance • Debates over in depth permeability reduction ability • Not for fractured or high permeability thief zones	• Pilot: in USA, China • Field applications in Argentina and Colombia. • In depth flow diversion
	PPG	• Millimeter-sized preformed particles • Swell up to 200 time by adsorbing formation water • Deformable when swelled • Pass through pore throat with various mechanisms	• Size and strength adjustable • High thermal and salinity resistance • Not affected by chromatographic separation, dilution, etc. • RPM properties	• Cannot be used for regular matrix • Possible wash out in fractures • Not effective for very high permeability fractures • Swelling can be limited at acidic condition	• Widely used in China both fractured and mature water flooded matrix reservoirs. • In depth flow diversion. • Fracture water shut off
	Microgel	• Micrometer-sized pre-cross-linked polymers • Size 0.3-2 µm • Reduce permeability with monolayer/multilayer adsorption on rock surface	• Various chemistry and properties • RPM property for water wet rocks at residual oil saturation • High thermal and salinity resistance	• High manufacturing and handling cost • Not for fractured reservoirs • Specific size adjustment required	• Field applications in Omani oilfields and China • Water shut off and in-depth flow diversion.
	TAP	• Small sized "Kernels" • 1-10 µm • Pop like "popcorn" up to 10 times with time and heat • Require a thermal front (cold water injection)	• Suitable for tight matrix thief zones • High temperature and salinity resistance • No injectivity issue and plugging near the wellbore	• Not for fractures or high permeability channels • Sensitive to acidic pH • Low Frr and possibility of washout	• Field application in Alaska, Brazil, Argentina, Gulf of Suez, Tunisia etc. • In depth flow diversion

Table 4-4: Comparison of the polymer gels based on the reservoir conditions (reference to information in table within the text).

Gel Category	Gel System	Reservoir Temperature	Salinity (TDS)	pH	Thief Zone Permeability
In-Situ Bulk	HPAM/Cr (acetate)	Up to 80 °C	Up to 30,000 ppm	5.5-7.5	Matrix and Fractures
	PAtBA/PEI	Up to 126 °C	Not given	> 8	Matrix and Fractures
Novel Microgels	CDGs	Up to 94 °C	Up to 35,000 ppm	Not given	Matrix
	PPGs	Up to 120 °C	Up to 300,000 ppm	> 6	> 0.3-1 D
	Microgels	Up to 140 °C	Up to 200,000 ppm	Not given	Matrix
	TAPs	20-150 °C	Up to 260,000 ppm	> 6	50-4000 mD

HPAM/Cr (III) acetate gel system with relative permeability modification properties is suitable for selective water shut off treatment of high permeability layers with crossflow and extended fractures from aquifer. This system can reduce the risk of damage to oil producing layers. PAtBA/PEI gel system, on the other hand, is useful for non-selective water shut off treatment. This system with addition of strength enhancer materials can provide sealing for near wellbore well integrity problems and also treatment of thief zones without crossflow. HPAM/Cr (III) acetate gel system application is limited to the reservoirs with temperature below 80℃ while PAtBA/PEI gel system with covalent bonding between polymer and cross-linker can withstand a higher temperature reservoir of 120℃.

Four different types of novel gels with various sizes from millimeter to sub-micrometer sizes were used for in-depth flow diversions. The application of these systems is restricted by their size, their mechanisms and thief zone permeability. CDGs were applied in field applications for in depth permeability reduction of ordinary permeability matrix reservoirs. However, the effectiveness of this technology to provide flow diversion is still point of controversy in the literature. PPGs were used for high permeability matrix reservoirs and fractures. Due to their relatively large size, these particles cannot be used for matrix reservoirs with low permeability. The application of PPGs for super conductive

fractures were also limited due to wash out of particles by post water flood. TAPs and Microgels with small sizes were used to provide flow diversion in matrix reservoirs with moderate permeability. In terms of reservoir temperature and formation water salinity, the CDGs systems application is limited to reservoirs with temperature up to 94℃ and salinity of 35,000 ppm. While PPGs, SMG microgels and TAPs were developed to withstand higher temperatures and salinities.

Based on the comparison of polymer gel systems provided in Table 4-3 and Table 4-4, parameters such as temperature, salinity and thief zone permeability of the reservoir could be used by the operators and reservoir engineers in the selection of proper polymer gel systems. This is because temperature and salinity of the reservoir strongly influence the overall effectiveness of the selected polymer gel systems. When the thief zone permeability is low the application of some polymer gel systems may not be effective. For example, in the application of PPGs, due to the large size of the particles, the propagation of the gel system in the reservoir is restricted by small pore throat size.

CHAPTER 5

Conclusions and Recommendations

5.1 Conclusions

In this study, a thorough review on the characteristics, development and field application results of six widely applied polymer gel systems for conformance control application in oil reservoirs was conducted. The study covered all six widely applied polymer gel systems that are not fully covered in the previous reviews. This study provides an updated review that covers the important properties of these polymer gels such as, gelation time, gel strength, gel stability, sealing ability, swelling capacity, relative permeability modification, selectivity of penetration and in-depth permeability reduction. Factor affecting these properties and selection of polymer gel systems for conformance control such as, temperature, salinity, pH, thief zone permeability and gel system composition are discussed in detail. The results of this study is helpful to the reservoir engineers and oil filed operators to choose the proper gel system based on environmental conditions such as temperature and salinity of the reservoir. Furthermore, the development, advancements, merits and controversies on these technologies as reported in recent laboratory experiments and field applications studies are provided. Finally, the comparison of these gel systems based on their advantages, disadvantages and their performance at different reservoir conditions are summarized.

These six systems include two conventional in-situ bulk gel systems: HPAM/Cr (III) acetate and PAtBA/PEI for water shut off and profile modification, and four novel gel systems: CDGs, PPGs, Microgels and TAPs for in depth flow diversion application.

- For conventional in-situ bulk gel, the main concerns were risk of damage to oil zone, fast gelation at high temperature, gel strength, gel stability and chromatographic separation of chemicals before gelation.
- HPAM/Cr (III) acetate gel system with relative permeability modification are suitable for selective water shut off treatment.
- To increase the gel strength, gel thermal stability, gelant composition control and delay gelation time of HPAM/Cr (III) acetate gel system at temperature above 80

^{o}C, various nanotechnologies such as cross-linker sequestration, nano-fly-ash and SiO_2 nano-composites were introduced.

- PAtBA/PEI gel system with controllable hydrolysis degree and high sealing ability can provide non-selective water shut off treatment at high temperature reservoirs.
- Various retarders were added to gel recipe to delay gelation time of PAtBA/PEI gel system at temperature higher than 126 ^{o}C.
- Strength enhancers, such as cement, silica flour and RSM were also used in field applications to improve the sealing ability and strength of this organically cross-linked system for water shut off applications.
- For in depth permeability reduction of fractures and matrix thief zones with various permeabilities, novel gels with millimeter to sub-micron meter size and different swelling capacities, thermal and salinity resistance were developed.
- CDGs were reported to be successfully used in the treatment of matrix thief zones at low to moderate temperature reservoirs. However, laboratory experiments contradict the ability of this system to provide in depth flow diversion.
- Microgels with various chemistry and properties were also introduced for matrix thief zones permeability reduction. However, due to the cost of manufacturing and handling of this system, economics of their application should be considered carefully.
- For high permeability matrix thief zones and moderated fractures, PPGs with high swelling capacity, high temperature and salinity resistance were successfully implemented.
- To prevent the washout of PPGs from extremely permeable fractures various methods such as, filling the gel pack with HPAM/Cr (III) gel, decreasing the gel pack permeability with using different particle size and re-assembling preformed particle gels were introduced in the lab experiments.
- To further improve the PPGs thermal stability and strength, nanocomposites such as nano fly ash, sodium montmorillonite and starch were introduced in the laboratory experiments.
- Temperature activated polymers (TAPs) with sub-micron size were introduced for in depth flow diversion of tight reservoirs with heterogeneity. For this system, the

mechanism of permeability reduction, applicable rock permeability and effect of surfactant need to be further studied.

5.2 Recommendations

Based on the results of the present study, the following recommendations for further research are suggested:

1) The retarders used to delay the gelation time of conventional in-situ bulks gels showed negative effect on the final gel strength. Therefore, new retarders with subtle impact on gel final strength of in-situ bulk gel should be developed.
2) Because the studies of nano-composites gel systems were restricted to their effects on gelation time or gel strength, future studies could focus on their effects on other properties of the gel systems such as relative permeability reduction, propagation into porous media and long-term thermal stability.
3) Preformed particle gels are not applicable for super conductive fractures. Therefore, new PPGs with higher swelling capacity and reassembling properties should be developed for plugging of extreme fractures.
4) Some laboratory experiments on propagation and permeability reduction of CDGs contradict the claimed benefits supported by field application results for the past two decades. The collaborative research is required to solve the long-lasting controversies.
5) The synergistic effect of surfactant on improve oil recovery of TAPs field applications have been questioned in the literature. Therefore, clarification of this issue could be the subject of future studies.

Mathematical models and simulation results for transport and rheological properties of polymer gel systems were beyond the scope of this study. However, understanding the propagation and permeability reduction of polymer gel systems in porous media can help the reservoir engineers to optimize the treatment. In this content, a thorough review of available mathematical models and simulators could be a future review target.

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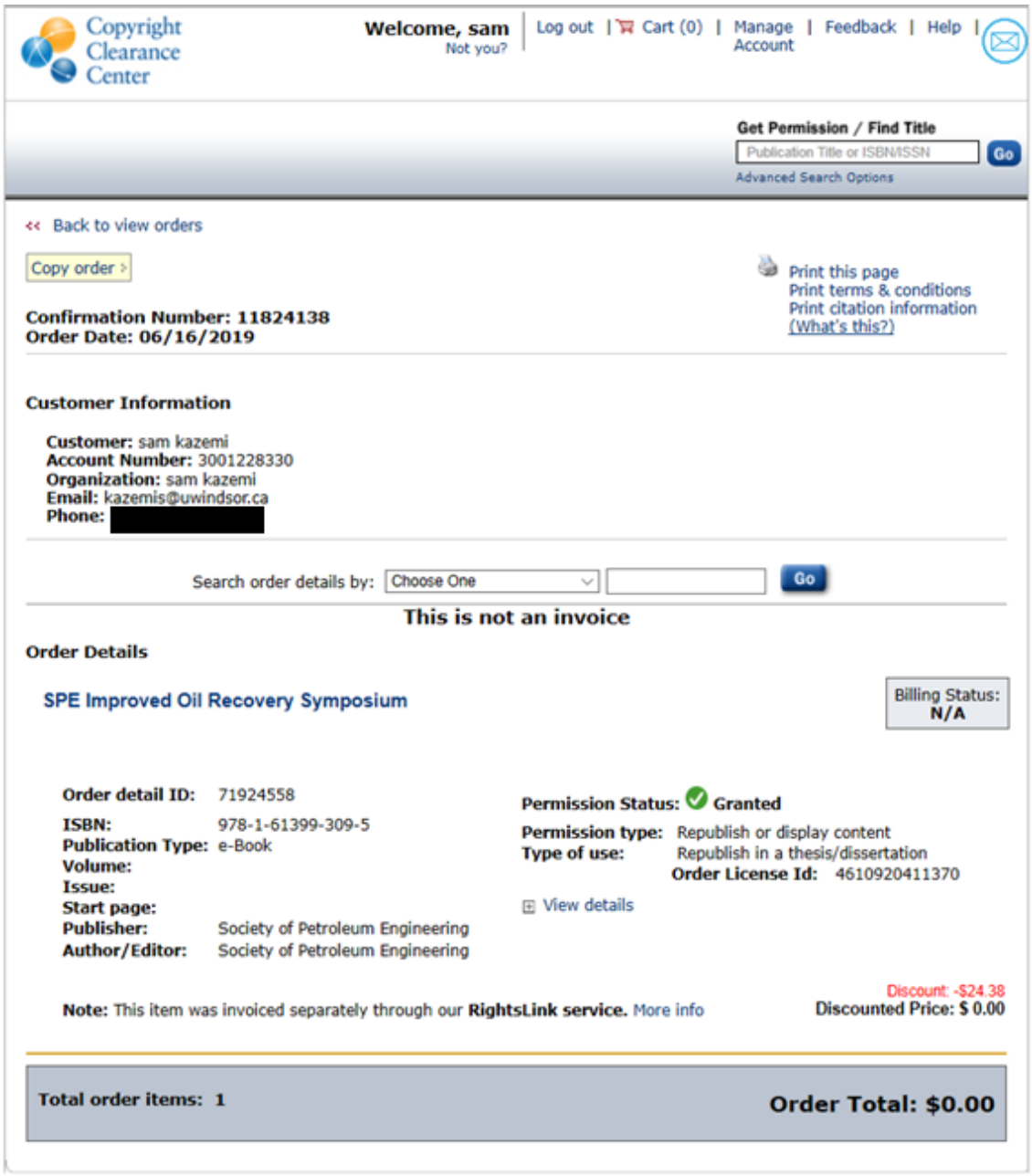

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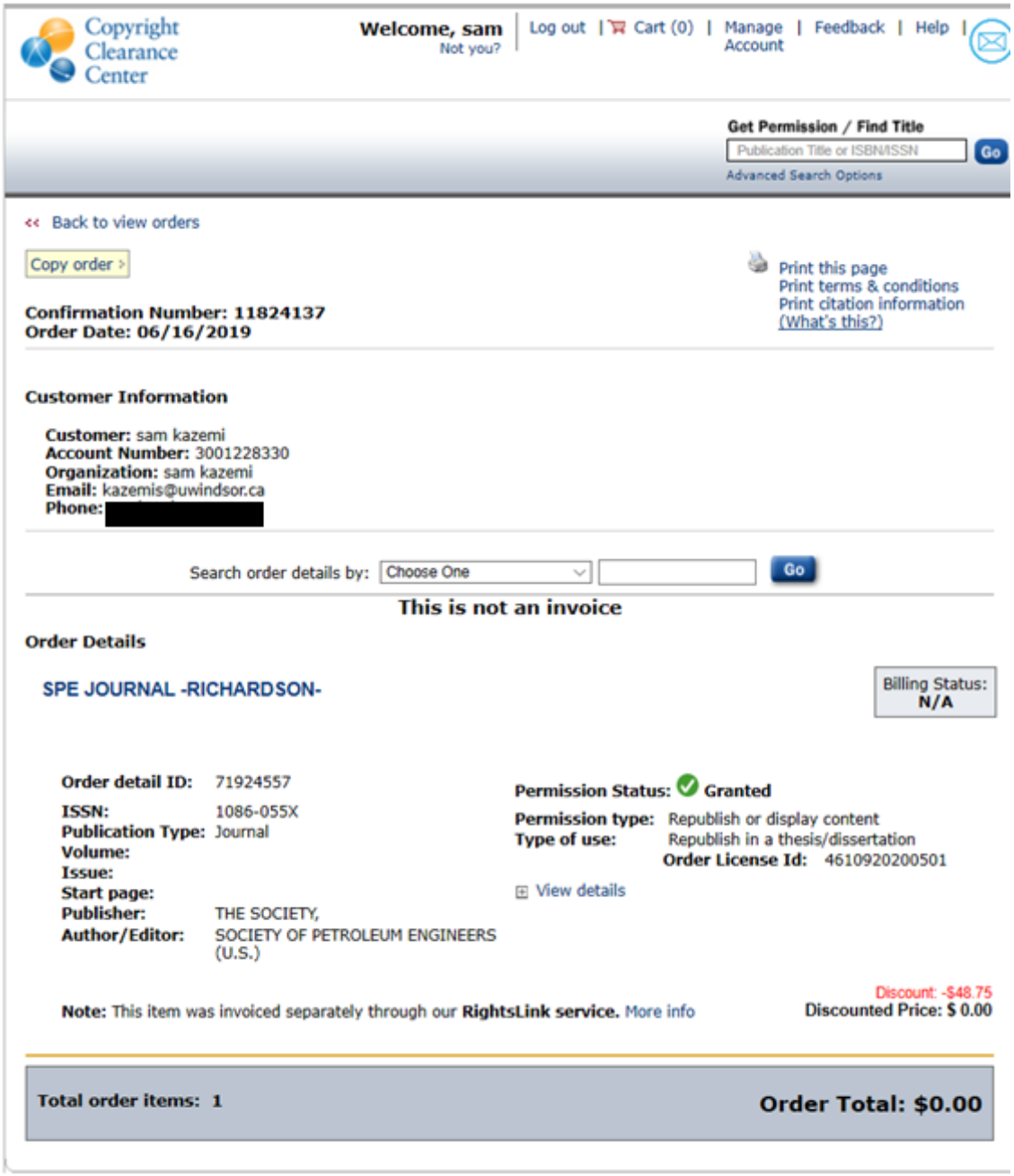

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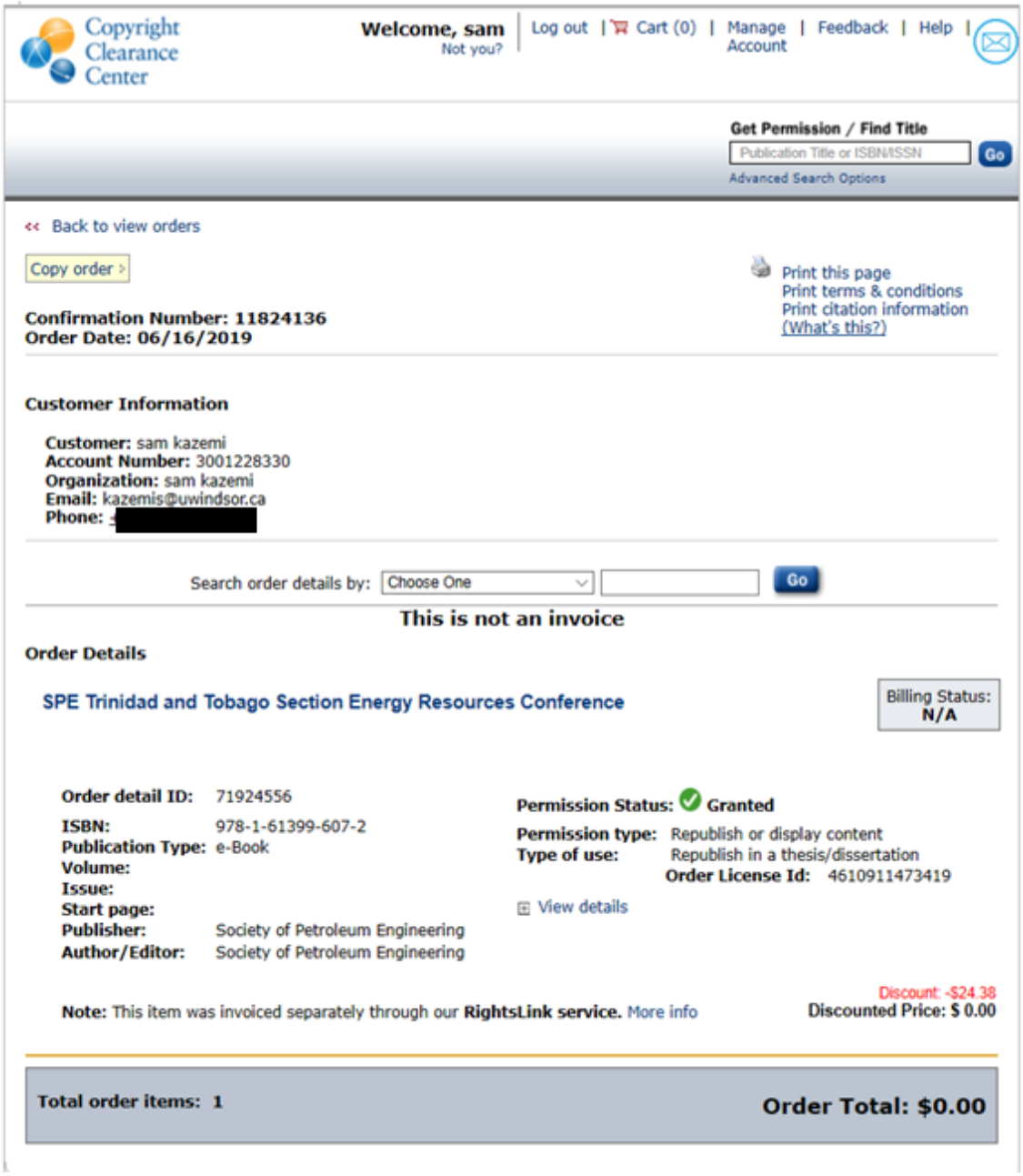

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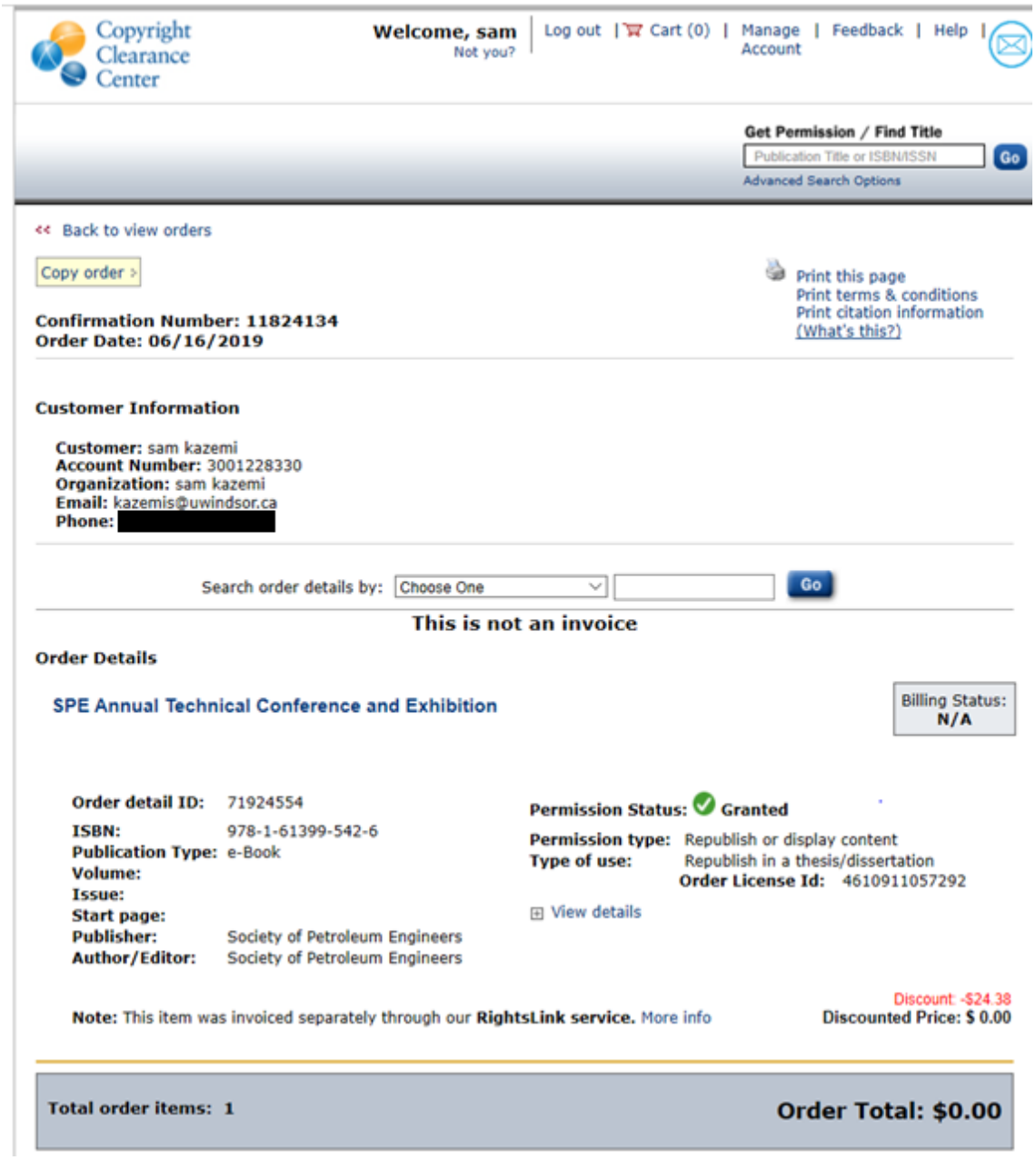

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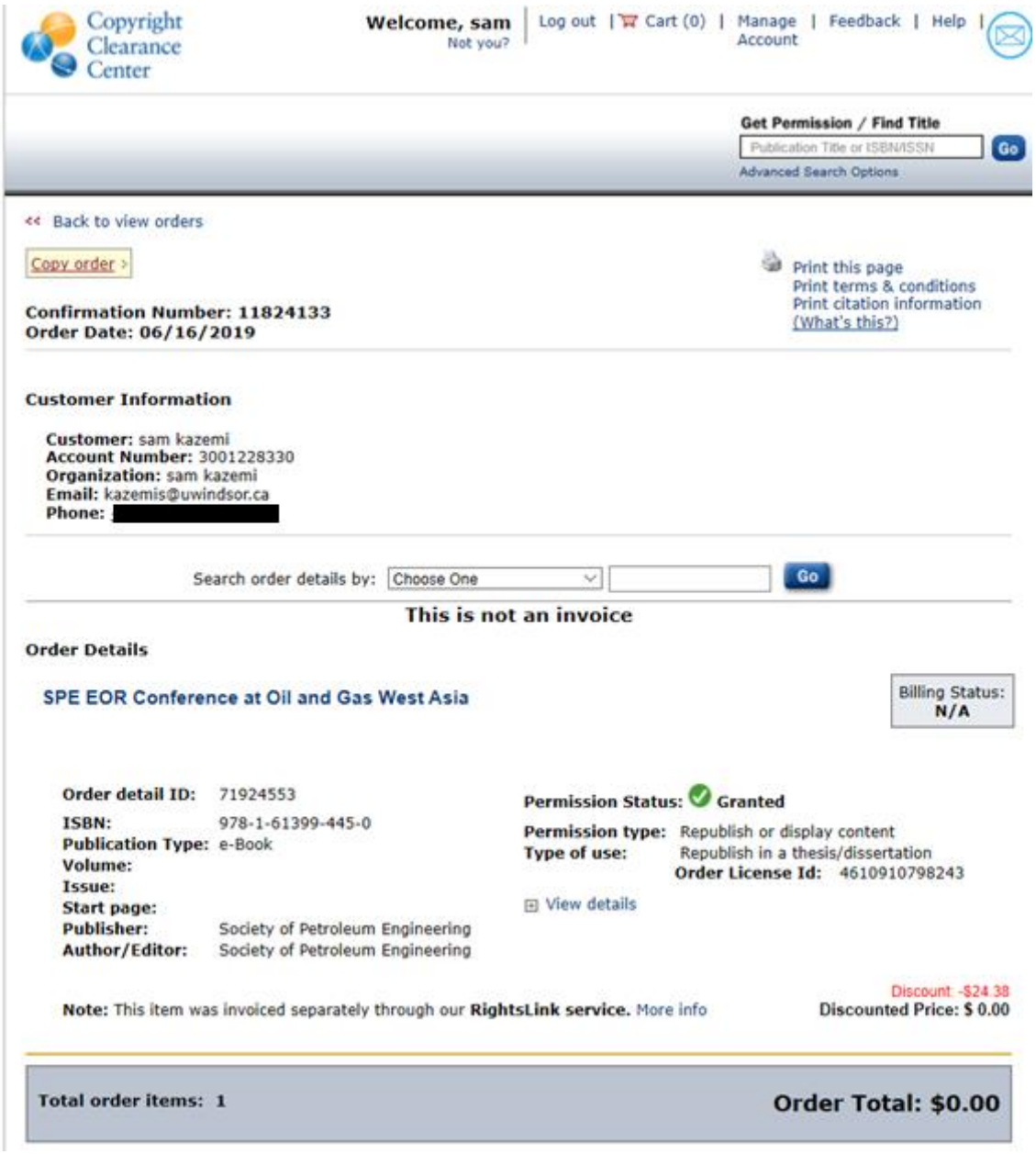

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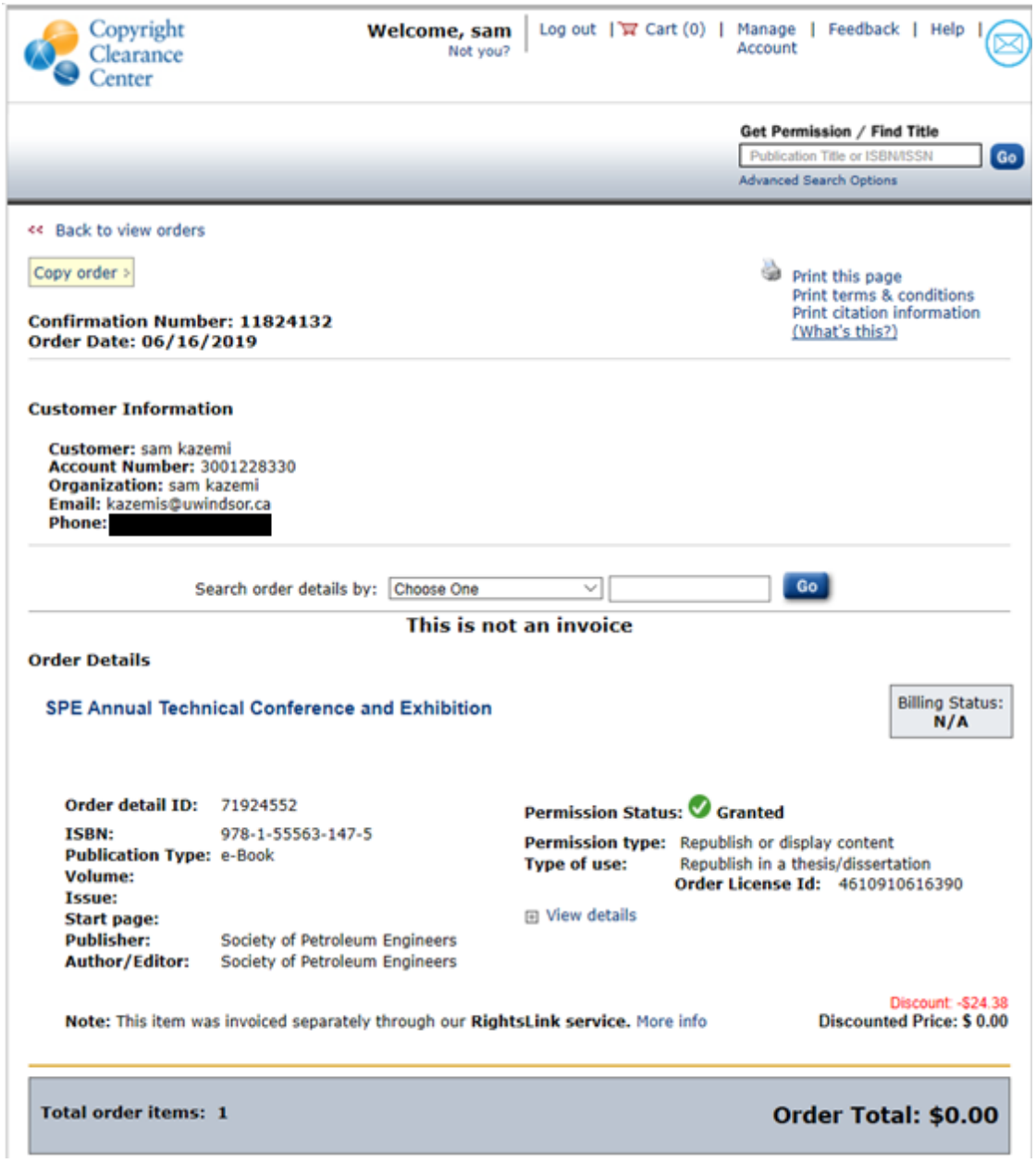

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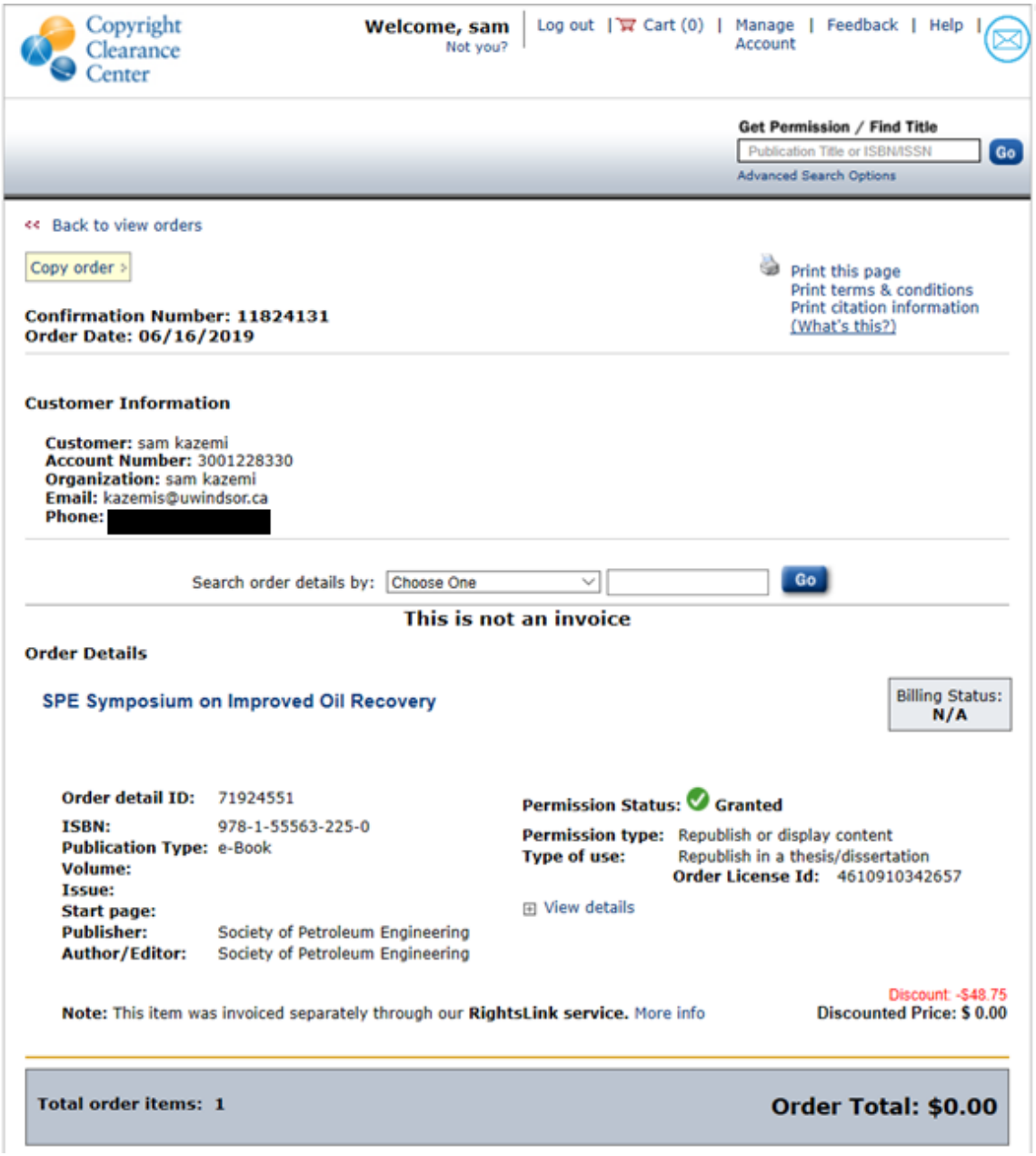

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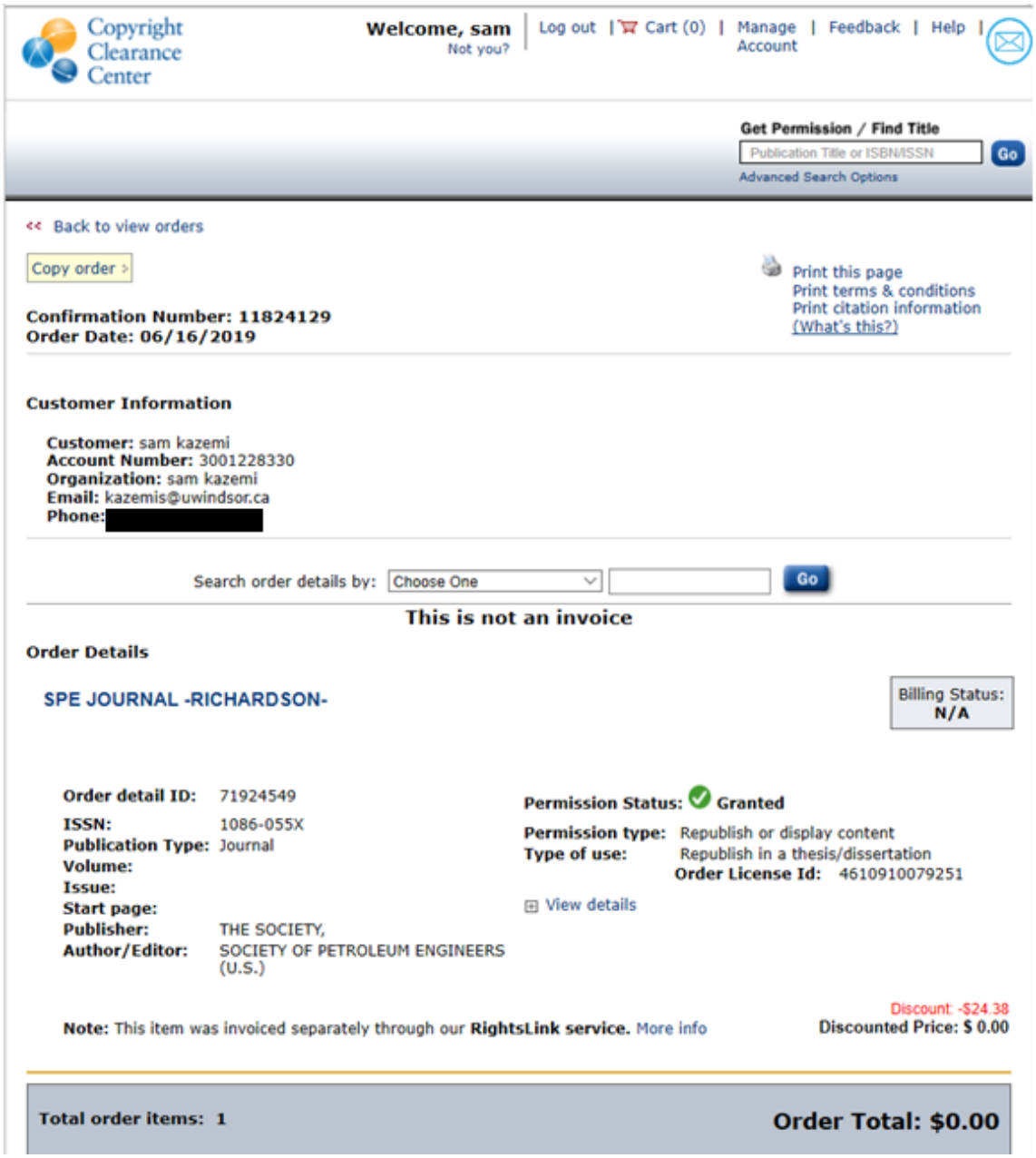

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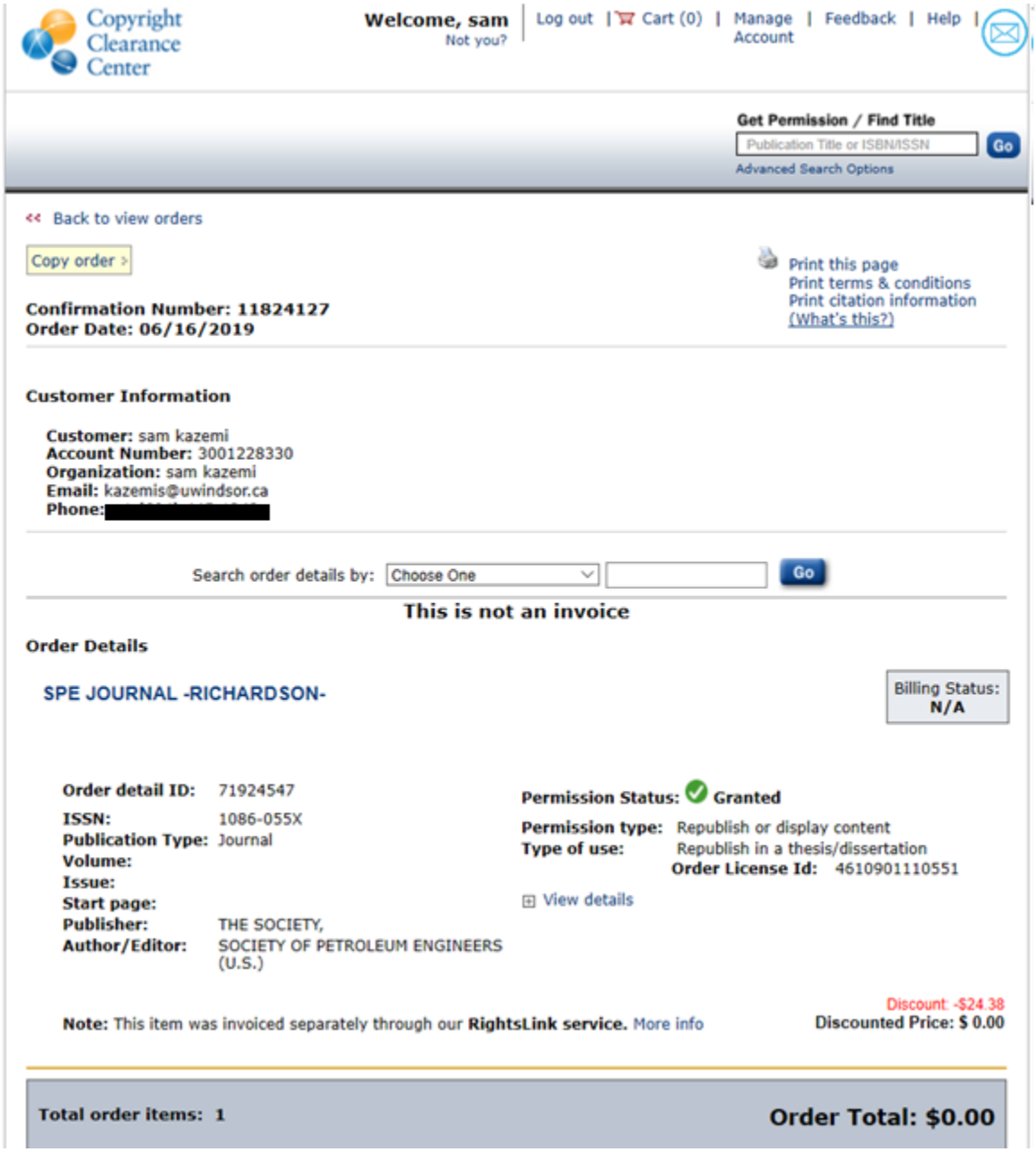

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Title of the article or chapter the portion is from	Preformed-Particle-Gel Transport Through Heterogeneous Void-Space Conduits
Editor of portion(s)	Abdulmohsin Imqam, Ze Wang, and Baojun Bai, Missouri University of Science and Technology
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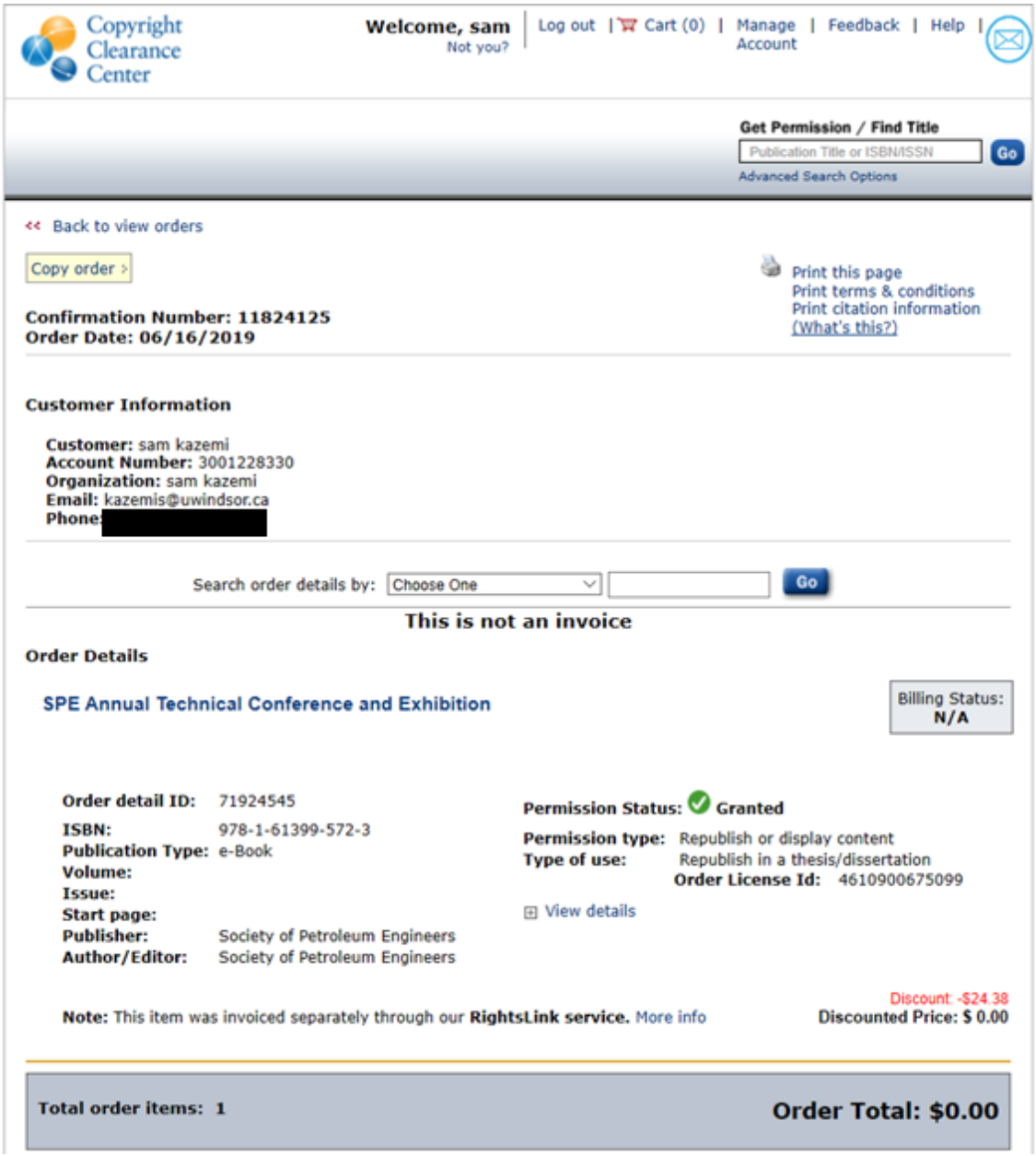

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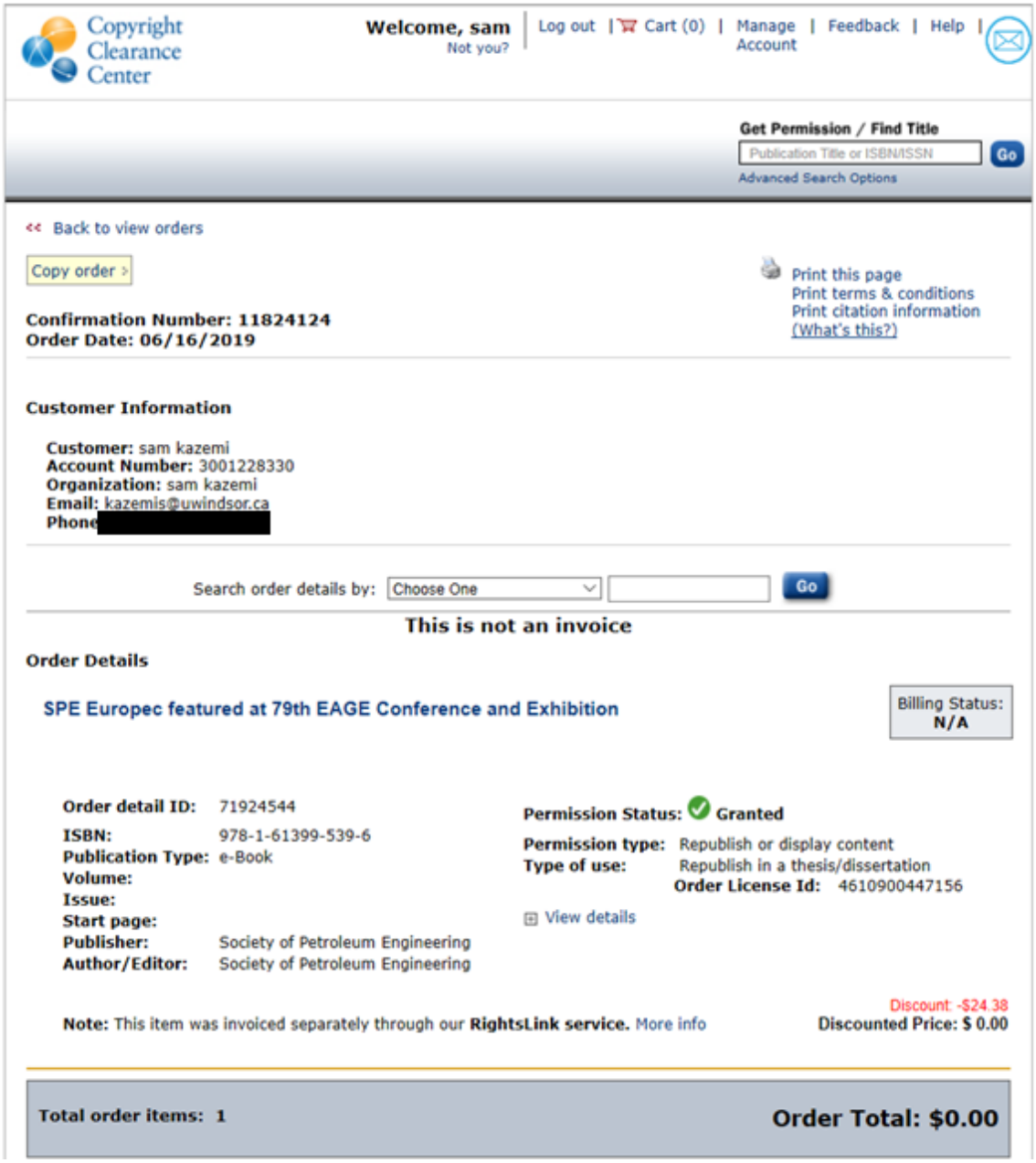

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Editor of portion(s)	Alain Zaitoun and Guillaume Dupuis, Poweltec
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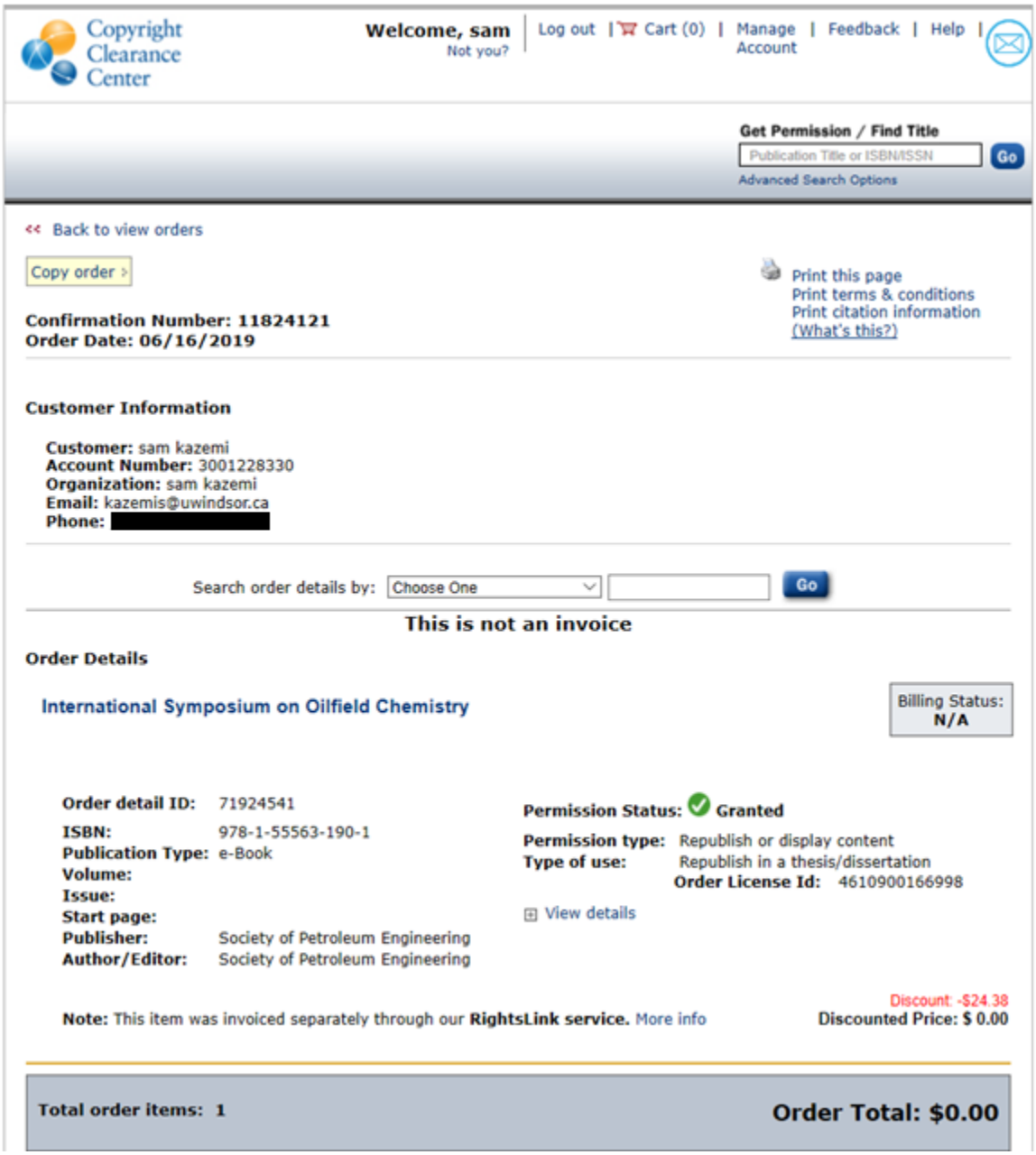

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Title or numeric reference of the portion(s)	Fig. 2 - Relative permeability modification by polymer adsorption
Title of the article or chapter the portion is from	Using Microgels To Shut Off Water in a Gas Storage Well
Editor of portion(s)	A. Zaitoun, SPE, R. Tabary, and D. Rousseau, Inst. Pétrole; T. Pichery and S. Nouyoux, Gaz de France; Français du and P. Mallo and O. Braun, Seppic
Author of portion(s)	A. Zaitoun, SPE, R. Tabary, and D. Rousseau, Inst. Pétrole; T. Pichery and S. Nouyoux, Gaz de France; Français du and P. Mallo and O. Braun, Seppic
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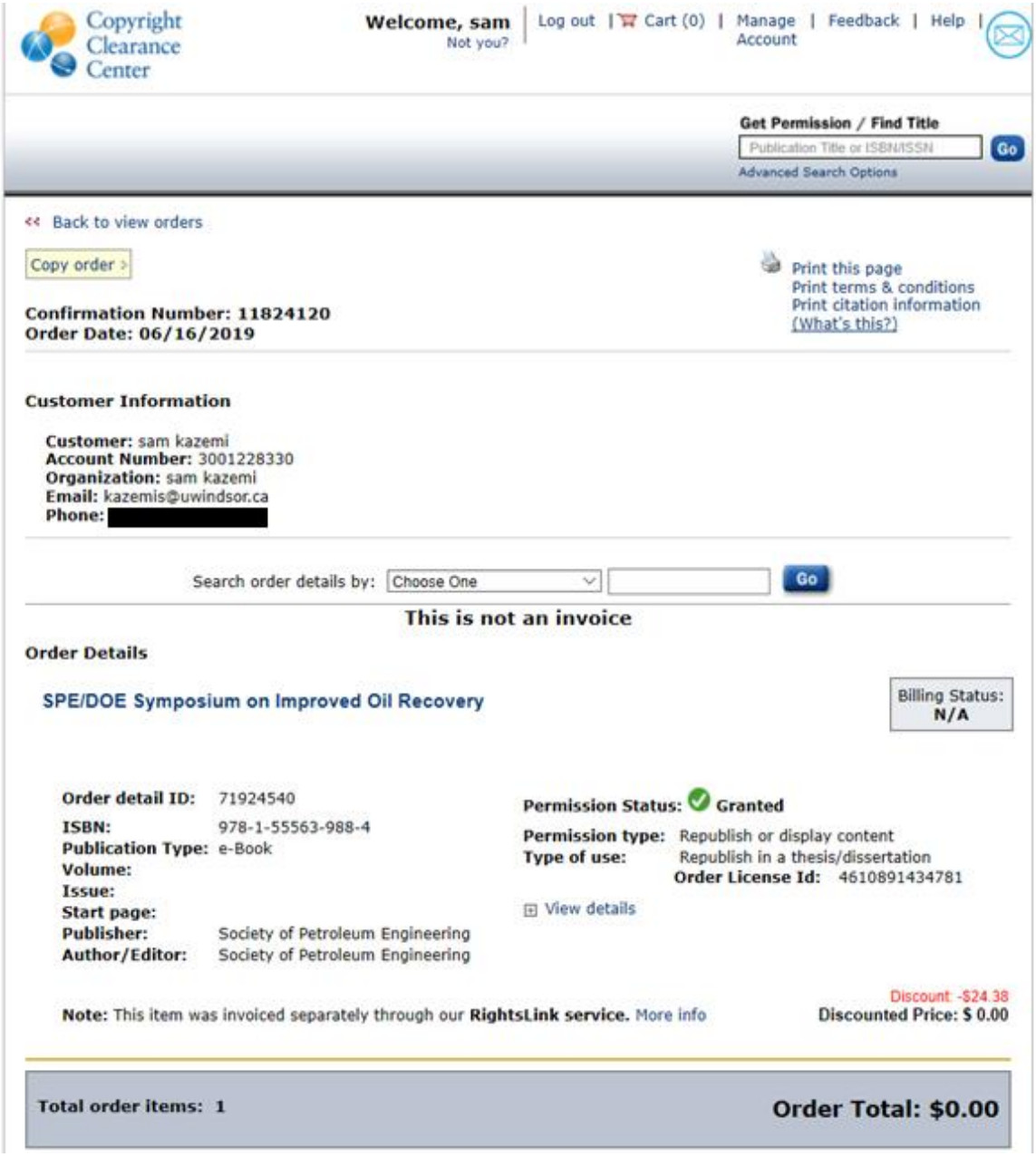

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Title or numeric reference of the portion(s)	Fig. 12 - Depth of penetration in lower permeability layer 2 and 3 (in cm) when microgels with a diameter of 1.5 µm have reached 100 cm in Layer 1.
Title of the article or chapter the portion is from	Disproportionate Permeability Reduction by Soft Preformed Microgels
Editor of portion(s)	Guy Chauveteau*, René Tabary*, Nicolas Blin*, Michel Renard*, David Rousseau* and Rien Faber**
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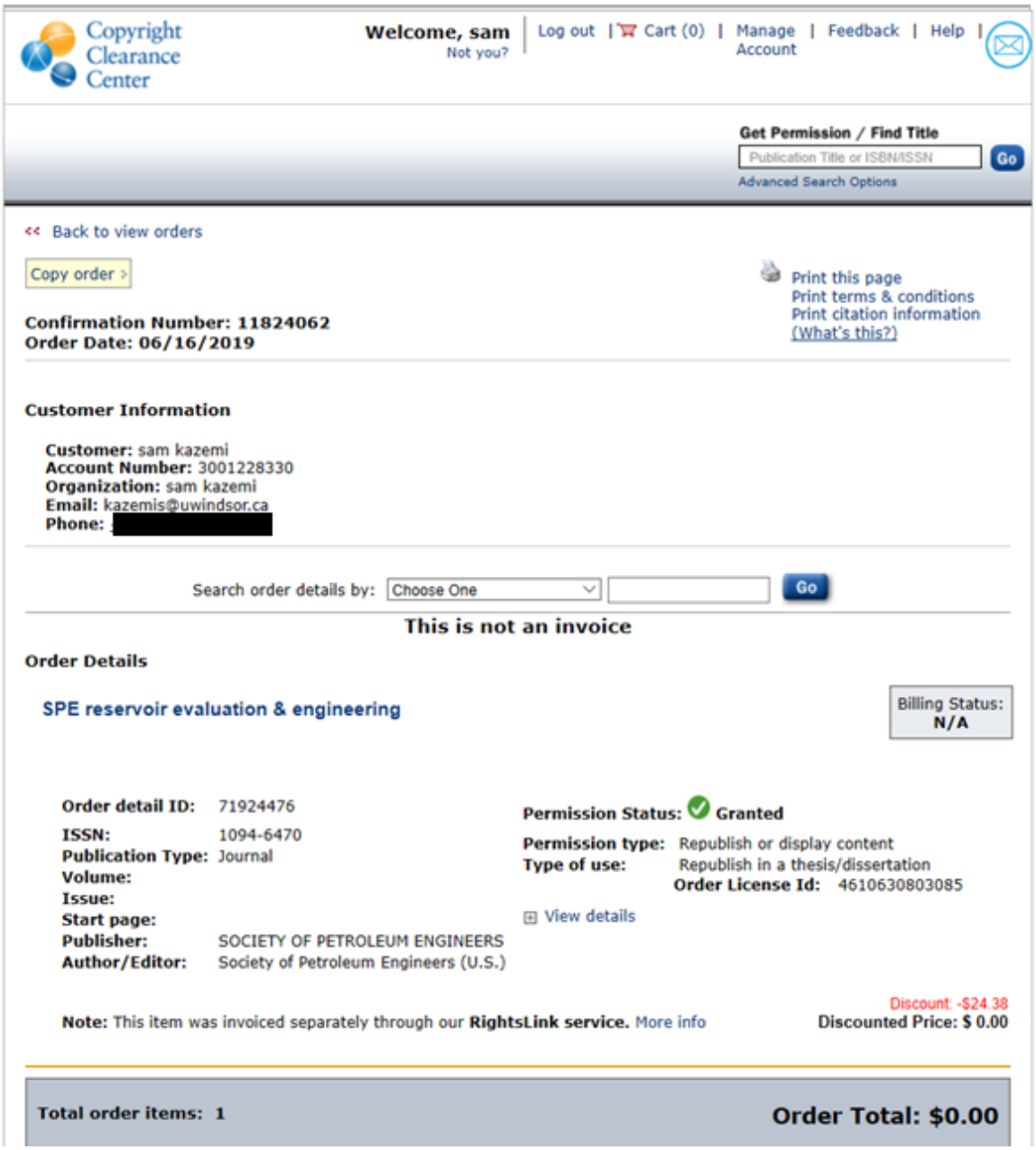

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Title or numeric reference of the portion(s)	Fig. 1. BrightWater is activated by heat and time. Particle sizes expand up to 10 times of its original size. Particles get adsorbed and retained by the rock and diverts flow into the unswept zones.
Title of the article or chapter the portion is from	Thermally Active Polymer to Improve Sweep Efficiency of Water Floods: Simulation and Pilot Design Approaches
Editor of portion(s)	Garmeh, R.; Izadi, M.; Salehi, M.; Romero, J.L.; Thomas, C.P. and Manrique, E.J., SPE, TIORCO
Author of portion(s)	Garmeh, R.; Izadi, M.; Salehi, M.; Romero, J.L.; Thomas, C.P. and Manrique, E.J., SPE, TIORCO
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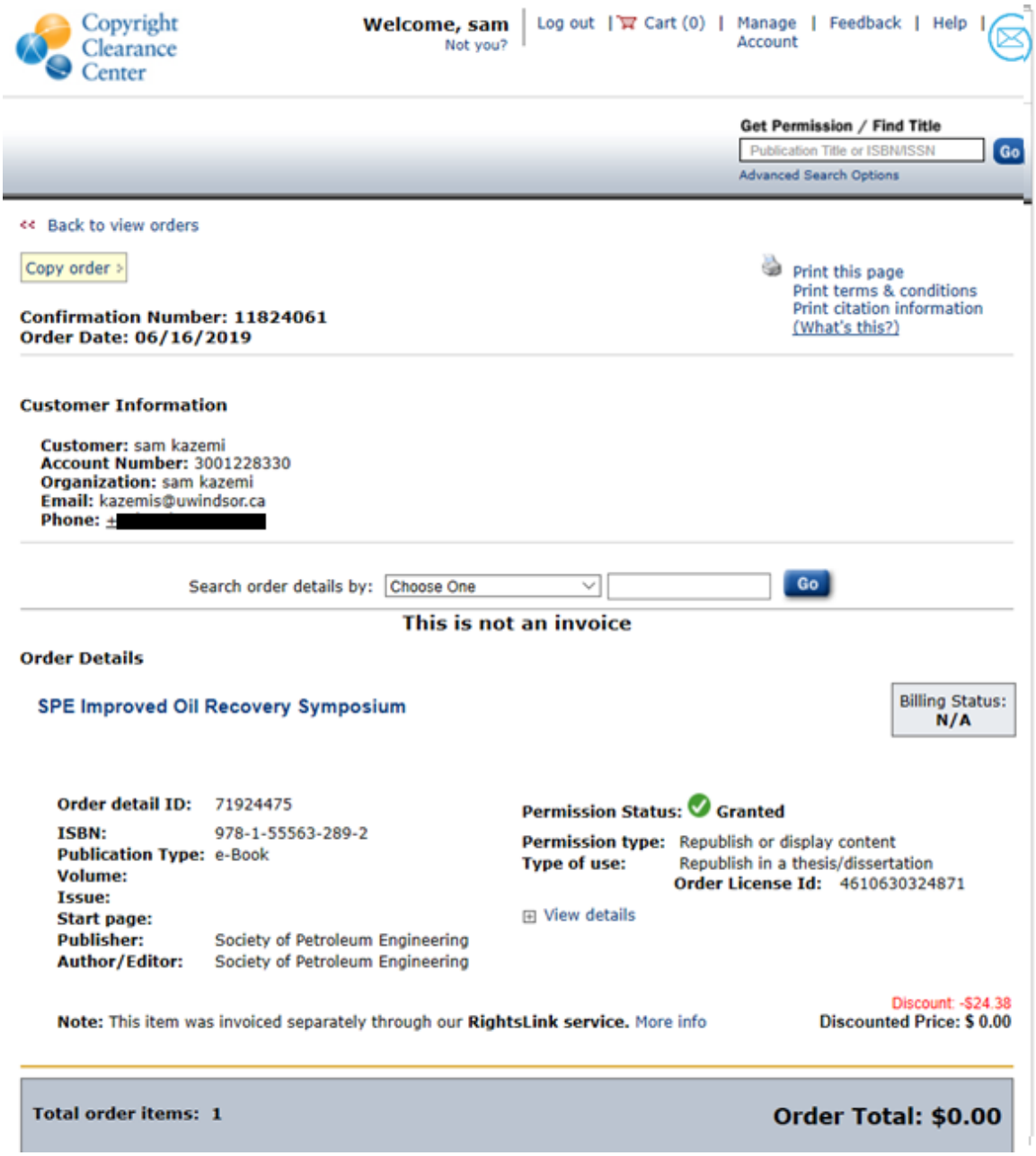

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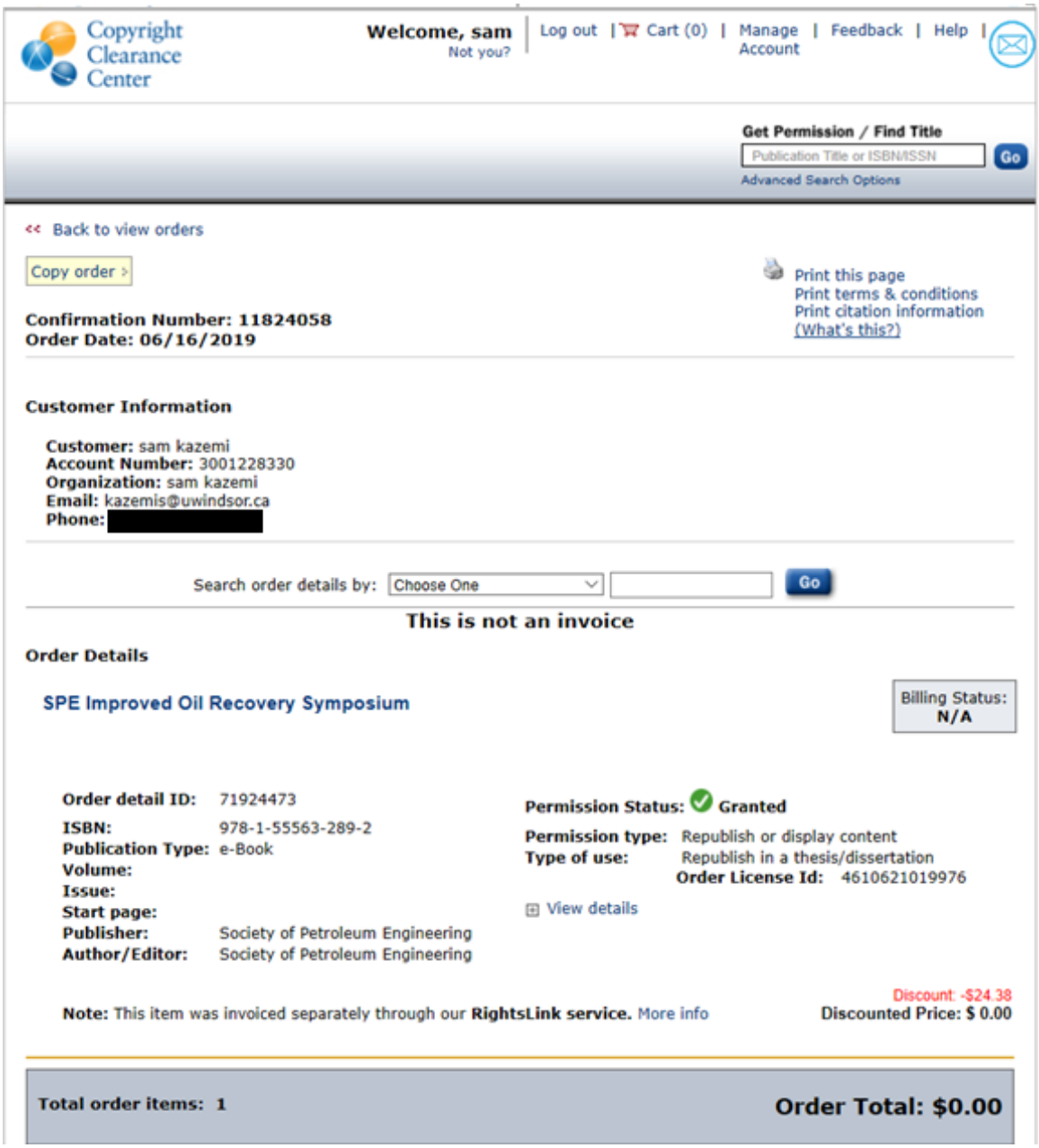

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Title or numeric reference of the portion(s)	Figure 6. Sand pack test results: 3000 ppm Bright Water Grade EC9378, KK injection water at 80° C.
Title of the article or chapter the portion is from	Deep Conformance Control by a Novel Thermally Activated Particle System to Improve Sweep Efficiency in Mature Waterfloods of the San Jorge Basin
Editor of portion(s)	J.L. Mustoni, SPE, Pan-American Energy; C.A. Norman, SPE, Tiorco; and P. Denyer, SPE, BP America
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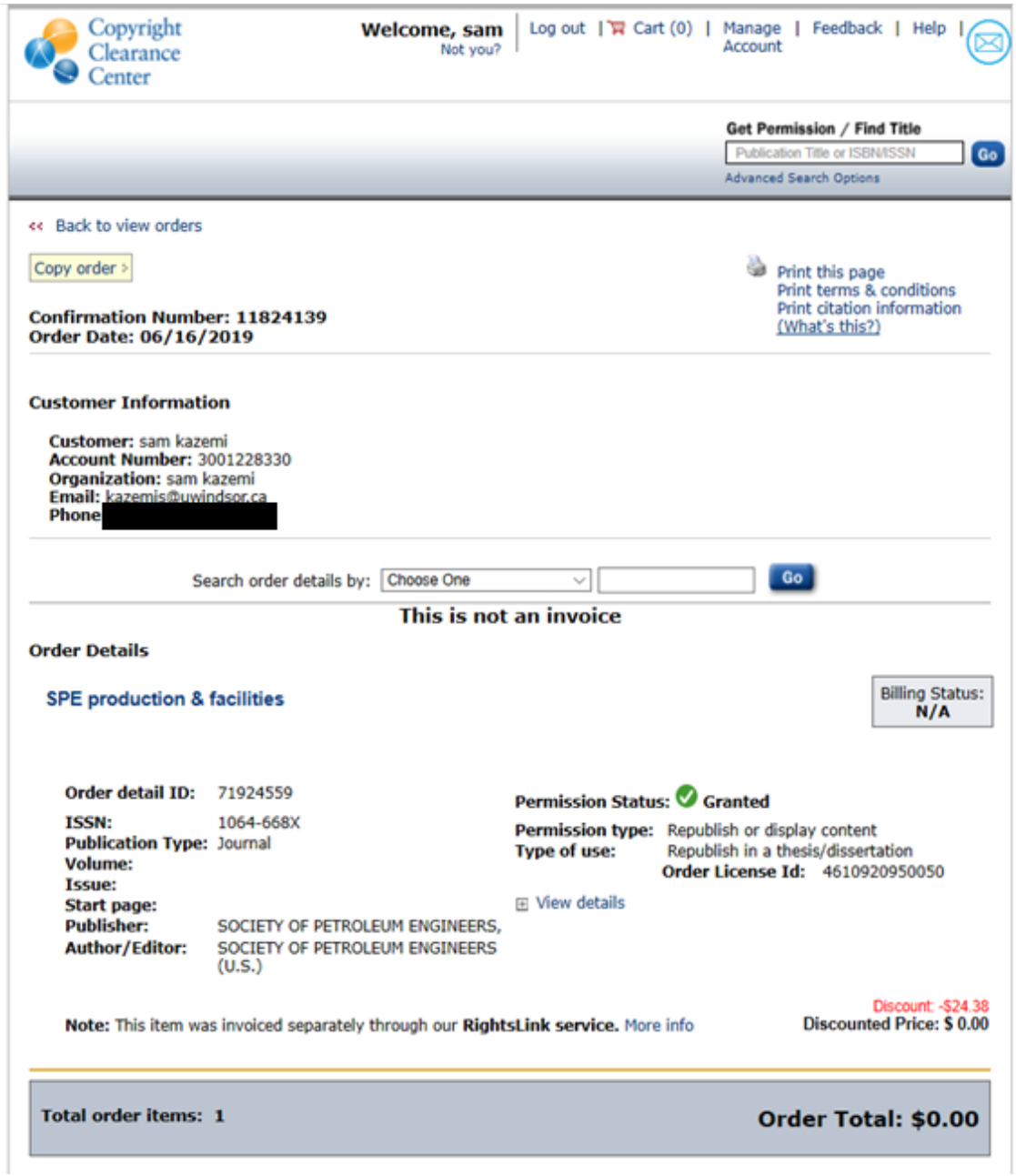

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Title of the article or chapter the portion is from	A Strategy for Attacking Excess Water Production
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Author of portion(s)	R.S. Seright, SPE, New Mexico Petroleum Recovery Research Center, R.H. Lane, SPE, Northstar
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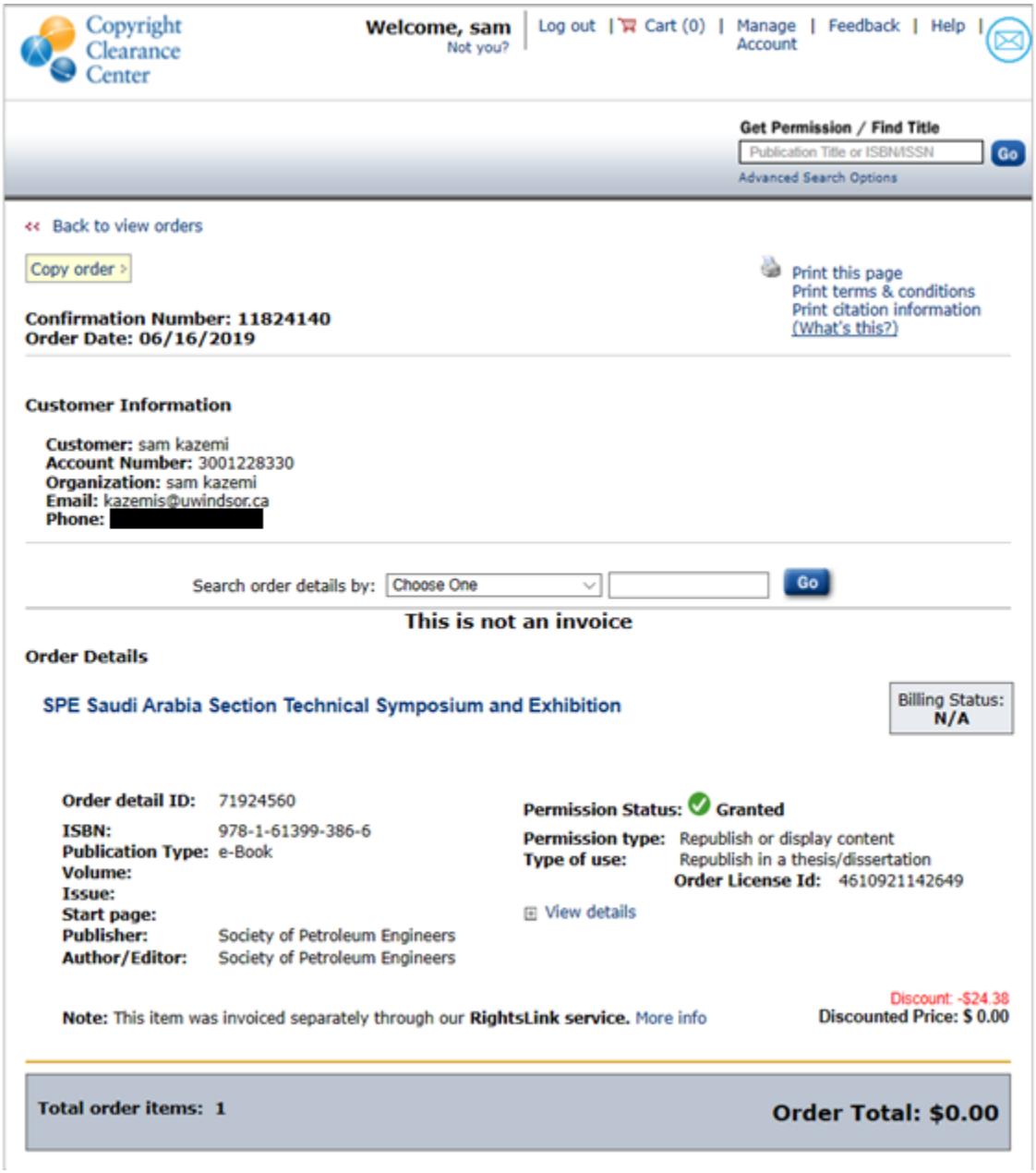

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Title of the article or chapter the portion is from	State-of-the-Art of In-Depth Fluid Diversion Technology: Enhancing Reservoir Oil Recovery by Gel Treatments
Editor of portion(s)	Ming Han, Amar J. Alshehri, Dimitrios Krinis and Stig Lyngra, Saudi Aramco
Author of portion(s)	Ming Han, Amar J. Alshehri, Dimitrios Krinis and Stig Lyngra, Saudi Aramco
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	University of Windsor, M.Sc., Windsor, ON, 2019